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LA-3366 (Rev)

Criticality Control in Operations with Fissile Material

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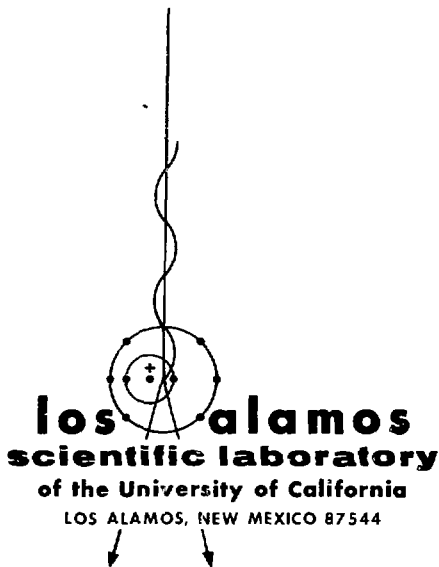


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Criticality Control in Operations with Fissile Material

by

H. C. Paxton

This report supersedes LA-3366.

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PREFACE

This account is intended to promote a broadened base for nuclear safety and to help provide a feeling for criticality control to anyone who works with fissile material.* As the quantity of reactor fuel increases, and as costs of fabrication, handling, and processing become more significant, it is presumed that restrictive rules by a few nuclear-safety specialists will no longer be tolerable. The alternative, which we espouse, is to make criticality control a live, active part of chemical and nuclear engineering, instead of a superposed topic with almost negative implications.

We assume that a feeling for nuclear safety can be developed without a working knowledge of theoretical reactor physics but with some appreciation for its capability. There are now considerable critical data, both experimental and computed, upon which empirical know-how can be based. This report emphasizes such data and attempts to make them understandable in terms of simple reactor-physics concepts.

We hope that a supplement eventually may give examples of application to a variety of real operations. Such illustrations are desirable to clarify methods of criticality control—further, they should add appeal for the process designer and operating engineer. There will be no attempt, however, to turn this into a handbook. Our purpose would be to show how the subject may be viewed and how one can go about incorporating nuclear safety into the design of an operation, not to provide a stereotyped set of rules.

Response to the original Los Alamos report of this title (LA-3366) was encouraging enough to stimulate the present revision. Among the many whose ideas are borrowed for this account, David R. Smith and Joseph T. Thomas have contributed especially generously. Further, Dixon Callihan, Elizabeth B. Johnson, and Joseph T. Thomas have kindly consented to the reproduction of numerous figures from the report TID-7028, "Critical Dimensions of Systems Containing U^{235} , Pu^{239} , and U^{233} ."

*For our purpose, "fissile" materials are the usual reactor fuels, ^{235}U , ^{239}Pu , and ^{233}U . The term "fissionable" refers to a broader class that includes, as well as these common fuels, other isotopes that can fission, e.g., ^{238}U , ^{240}Pu , and ^{232}Th .

CRITICALITY CONTROL IN OPERATIONS

WITH FISSILE MATERIAL

by

H. C. Paxton

ABSTRACT

This discussion of criticality control is intended to encourage a working knowledge on the part of those who design and perform operations with fissionable material. As background, requirements of the Atomic Energy Act are interpreted, and nuclear-safety experience is outlined. Both are shown to be compatible with reasonable principles of nuclear safety. Next, empirical criticality information is presented to help develop a feeling for conditions to be avoided during operations. Criticality-control methods that are consistent with the stated principles and available criticality data are described in the final section.

I. BACKGROUND

THE AEC

We cannot discuss nuclear safety realistically without examining the influence of the U. S. Atomic Energy Commission. The AEC was appointed legal guardian of the nuclear industry by Congress through the Atomic Energy Act. Like many a parent, it became accustomed to nursing the infant industry, and now has some difficulty adjusting to the problems of an adolescent. Failure to relax early controls retards the development of responsibility, yet the youth may get into trouble if there is relaxation before responsibility is demonstrated. This apparent paradox will be resolved only when the industry recognizes its own maturity and the AEC does also. Even then, a dual responsibility traces back to the Act, and satisfactory balance of this responsibility will depend upon sympathetic mutual understanding.

The basis for this understanding must start with the safety responsibility that Congress requires of the AEC. The tone is set by typical quotations from the Act.

Sec. 3. PURPOSE: "It is the purpose...to effectuate the policies...by providing for...a program to encourage widespread participation in the

development and utilization of atomic energy for peaceful purposes to the maximum extent consistent with the common defense and security and with the health and safety of the public..."

Sec. 31. RESEARCH ASSISTANCE: "...the Commission is authorized and directed to make arrangements ...for research and development activities relating to...the protection of health and the promotion of safety during research and production activities."

Sec. 41. OWNERSHIP AND OPERATION OF PRODUCTION FACILITIES: "Any contract...shall contain provisions...obligating the contractor...to comply with all safety and security regulations which may be prescribed by the Commission."

Sec. 53. DOMESTIC DISTRIBUTION OF SPECIAL NUCLEAR MATERIAL: "Each license...shall be subject to the following conditions...special nuclear materials shall be distributed only pursuant to such safety standards as may be established by rule of the Commission to protect health and to minimize danger to life or property...."

Sec. 182. LICENSE APPLICATIONS: "...the applicant shall state...such...information as the Commission may, by rule or regulation, deem necessary

...to find that the utilization or production of special nuclear material...will provide adequate protection to the health and safety of the public."

Of course, these provisions are subject to interpretations ranging from stringent to reasonably liberal. For example, the word "adequate" in the last quotation may be ignored or emphasized. Strict interpretation was natural during the infancy of the nuclear industry, but liberalization should be expected as the industry matures and demonstrates its responsibility. A reasonable attitude toward safety regulation is expressed in commentaries on the Act that appear in the document "Improving the AEC Regulatory Process," dated March 1961, which was prepared by the staff of the Joint Committee on Atomic Energy, under James T. Ramey, then Executive Director. The practical attitude is illustrated by a statement (p. 61) about safety in achieving atomic goals.

"The primary objective of the atomic energy regulatory process should be, of course, to protect the health and safety of the public and employees in industrial and other uses of radiation. As noted earlier, absolute safety is not the objective, however, for this would require discontinuance of all nuclear development. Therefore, national goals, such as development of nuclear weapons, long-range space exploration through use of nuclear propelled vehicles, achievement of economic nuclear power, increased use of radioisotopes, and pursuit of basic atomic research, must be considered in determining the reasonableness of safety requirements."

As recognized in this statement, no processing of fissile material presents zero risk.

In summary, the Act requires both contractor and licensee to comply with AEC regulations designed "to protect health and to minimize danger to life and property" or to "provide adequate protection to the health and safety of the public." These regulations are supposed to recognize "widespread participation in the development and utilization of atomic energy for peaceful purposes to the maximum extent consistent" with the above safety aims (and with the common defense and security). Furthermore, the AEC is directed to arrange for technical safety guidance and safety promotion.

The resulting overall picture of safety regulation is fluid, something that adjusts to technical knowledge, instead of arbitrary requirements that

are fixed for all time. This is important, because it permits us to view criticality control within its technical bounds, not within the limitations of existing or proposed regulation. Furthermore, it appears that the way is left open for the mutual understanding that was mentioned earlier. To bring about complete understanding, the AEC must keep abreast of technical developments and objectives of the nuclear industry, and licensees and contractors must demonstrate their competence and contribute to the improvement of regulations.

SAFETY EXPERIENCE

There have been six supercritical accidents in chemical process equipment but none associated with mechanical processing, storage, or transportation.* All occurred with aqueous solutions; four involved highly enriched uranium, and two involved plutonium. Two of the excursions took place in areas that were shielded to accommodate irradiated fuel, so that personnel were protected from direct radiation.

The results of these 6 accidents have been 2 deaths, 19 significant overexposures to radiation, no equipment damage, and negligible loss of fissile material. In no case was there any danger to the general public.

Each incident was a result of process or equipment difficulty or maloperation (generally a combination). There was no contribution by faulty criticality information, nor by error in its interpretation.

Before proceeding from these general remarks to more specific features of the accidents, it may be useful to picture the usual characteristics of a supercritical excursion in a solution. Typically, there is a fission spike that is terminated by heating and consequent thermal expansion of the solution and by bubble formation. If there is no loss of material, as by splashing, fissioning continues at a reduced rate that may have less intense spikes than the first as bubbles sweep out of the solution. Continued addition of solution after the initial burst will amplify these secondary spikes. Of course, loss of solution, or redistribution of material may terminate the reaction after the initial burst.

Numbers of fissions, which are quoted in the following accounts, may require translation into

* We overlooked an instance of accidental criticality reported by J. T. Daniels, H. Howells, and T. G. Hughes: "Criticality Incident—August 24, 1970, Windscale Works," *ANS Trans.* 14, No. 1, 35-36 (June 1971).

more familiar terms. A modest burst of 3×10^{16} fissions deposits 1 MW-sec, 240 kcal, or 950 BTU of energy, most of which heats the solution.

A complete listing of criticality accidents appears in a review by W. R. Stratton,¹ and details are given in the references he cites. Although we will confine our attention to accidents in processing plants, conditions that have led to excursions in critical facilities are also instructive. The following accounts of plant accidents are intended to provide not only an idea of the consequences but a general introduction to nuclear-safety practices.

The Y-12 Plant, Oak Ridge, Tennessee—June 16, 1958. This accident occurred in an area for the recovery of U(93)* from scrap, while a material inventory was in progress. An empty solution-storage cylinder (geometrically safe—see Sec. III) was being cleaned, and wash water that flowed into a 55-gal drum led to the excursion. Solution had leaked into the cylinder (during the time between emptying and washing) through a valve that was supposed to provide isolation from other process equipment still in operation. Concentrated solution that first flowed into the large-diameter drum was too shallow for criticality until diluted by some of the wash water that followed. Initial criticality occurred with about 2.1 kg of ^{235}U in 56 liters of solution. Further dilution ultimately reduced the uranium concentration enough to make the system subcritical, but not until a succession of bursts had produced a total of 1.3×10^{18} fissions in 3 min. Because of the relatively low flow rate, it is estimated that only 10^{16} fissions occurred in the first and largest burst. This is consistent with the observation that the reaction was not violent enough to splash solution out of the drum. An initial "blue flash" was reported.

One man who was about 6 ft from the drum received an exposure of 461 rem; other exposures were 428 rem at 18 ft, 413 rem at 16 ft, 341 rem at 15 ft, 298 rem at 22 ft, 86 rem at 31 ft, 86 rem at 37 ft, and 29 rem at 50 ft. Exposures and distances from the drum do not correlate closely primarily because some routes taken out of the plant were more favorable than others. The exposures resulted from little more than the initial burst (from which there is no

escape), because radiation alarms signaled the accident, and the area was evacuated promptly. The importance of rapid departure can be appreciated by comparing actual exposures with the 400-to 500-rem range within which the chance of survival is estimated to be about 50%.²

The following measures were subsequently adopted to prevent similar accidents: Equipment is isolated by actually disconnecting transfer lines that may contain fissile material. Only containers that would be safe for U(93) solutions are permitted in process areas (e.g., waste baskets are perforated, and mop buckets have been replaced by geometrically safe containers).

The Los Alamos Scientific Laboratory—December 30, 1958. This accident involved equipment for treating dilute raffinate from a plutonium recovery plant. Residual plutonium (supposedly ~0.1 g/liter) and small quantities of americium were recovered from the raffinate by solvent extraction in large tanks. Again, a material inventory was in progress, and the tanks (all closed) were to be emptied and cleaned, one by one. Presumably to simplify this process, residual materials and nitric-acid wash solutions from four vessels were emptied into one, a vertical 225-gal, 38-in.-diam tank. This collection was made possible by the existence of many interconnecting transfer lines. The excursion occurred in this tank when its stirrer was turned on. Investigation showed that there was 3.27 kg of plutonium in an 8-in.-thick organic layer (160 liters) that floated on a dilute aqueous solution (60 g of plutonium in 330 liters). The initial action of the stirrer was to thicken the center of the organic layer enough to make it supercritical. Continued stirring immediately established a vortex, then mixed the organic and aqueous phases, diluting the plutonium enough so that criticality did not recur. The excursion consisted essentially of a single spike of 1.5×10^{17} fissions. The operator, who was standing against the tank while looking into a sight glass, received an exposure of 12,000 rem ($\pm 50\%$) and died 36 h later. Two men who went to help the victim received exposures of 134 rem and 53 rem. There was no damage to equipment and no contamination, although the shock displaced the tank support 3/8 in. and knocked the operator off a small ladder. A radiation alarm 175 ft away was activated,

*U(93), for example, means uranium whose ^{235}U enrichment is 93 wt%.

and a flash of light accompanying the excursion was seen from an adjoining room.

The only explanation found for the presence of 3.3 kg of plutonium in this process which had an expected inventory of 0.125 kg is a gradual accumulation of solids during the 7½-yr history of operation. The entire recovery plant had been scheduled to be rebuilt after another 6 months of operation. Instead the old equipment was retired immediately.

Apart from conversion to safer equipment, the following practices were adopted as a result of the accident. Written procedures for all operations and for emergencies were improved, and emphasis on nuclear-safety training was increased. Gamma-sensing radiation alarms were designed and installed to provide complete coverage of process areas. Solution-transfer lines not required for a specific operation were blocked to minimize the opportunity for abnormal interchanges. Neutron "poison" in the form of cadmium-nitrate solution was placed in vent tanks and vacuum-buffer tanks to protect against accidental introduction of plutonium. (Borosilicate glass Raschig rings have been used for this purpose in some plants.) Furthermore, periodic surveys with portable neutron detectors are conducted to detect abnormal deposits of plutonium.

The Idaho Chemical Processing Plant, National Reactor

Testing Station—October 18, 1959. The primary function of the Idaho Chemical Processing Plant is to purify and concentrate the fissile material in spent reactor fuel. Thick concrete shielding protects personnel from exposure to the highly radioactive fuel. The excursion occurred as the result of air sparging of a bank of safe storage cylinders that contained U(93) solution (170 g ^{235}U /liter). The sparging initiated a siphoning action that transferred about 200 liters of solution (34 kg ^{235}U) from the storage cylinders into a 5000-gal tank containing about 600 liters of water. Criticality in this tank led to a total of 4×10^{19} fissions during perhaps 20 min. It is guessed that a power spike of about 10^{17} fissions was followed by smaller spikes, and then by more-or-less stable boiling of the solution. The reaction terminated after an estimated 400 liters of water was distilled into another tank.

Although there was no direct neutron and gamma exposure, gaseous and air-borne activity spread into operating areas through vent lines and drain

connections and triggered radiation alarms. Significant beta-radiation dosages, 50 and 32 R, were received by only two persons during plant evacuation. Again, no equipment was damaged.

The desirability of a valve in the line through which solution was transferred to the 5000-gal tank had been foreseen, and action to correct this deficiency had begun. The incident uncovered the need for improved evacuation procedures and demonstrated the usefulness of radiation alarms in areas that might be effected by a nuclear incident occurring elsewhere. Equipment and operating procedures were reviewed to establish several lines of defense against inadvertent transfers of fissile material. The Idaho Chemical Processing Plant, National Reactor Testing Station—January 25, 1961. This incident differs from the others in that there is considerable justification for viewing it as only a minor deviation from normal operations. Heavy concrete shielding protected personnel from direct radiation, the ventilation system prevented airborne activity from entering work areas, and equipment design was such that there was no practical possibility of a destructive or persistent excursion. We discuss the incident to illustrate a situation that constituted no hazard, but which could have had serious consequences had there been no shielding.

The excursion occurred when about 40 liters of uranyl-nitrate solution (200 g U(93)/liter) was forced upward from a 5-in.-diam section of an evaporator into a 24-in.-diam vapor-disengagement cylinder that was above the normal solution level. Presumably air had been introduced into associated lines during attempts to clear a plugged line and to improve the operation of two pumps. When the bubble of air reached the evaporator, solution was expelled from the lower section. The excursion, probably a single spike, had a magnitude of 6×10^{17} fissions. Although radiation was sufficient to trigger alarms and cause evacuation of the plant, no personnel exposure was greater than 100 mr.

Because the possibility of an excursion in the vapor-disengagement cylinder had been foreseen, lines at its base led to two geometrically favorable vessels with provisions for overflow to the floor. This arrangement, as well as other features, prevented both a large pressure buildup and a sustained reaction. Largely to avoid the embarrassment of a

recurring incident, a grid of stainless steel containing 1% boron has been installed in the 24-in.-diam cylinder to "poison" any solution that might enter. There are also added precautions against the introduction of air into any solution lines where its effect could be undesirable.

The Recuplex Plant, Hanford, Washington—April 7, 1962. The multipurpose Recuplex facility, for plutonium-recovery operation, started as a pilot plant in 1955, but with successive changes became a production facility. The various portions of this versatile plant were contained in room-size plastic hoods (gloveboxes) to prevent external contamination. A thorough cleanup, necessitated by deterioration of equipment and resulting leakage, was near completion at the time of the accident. Even visibility through the plastic walls of the hoods had become poor.

The 69-liter glass tank in which the excursion occurred was normally used for transfer of a dilute side stream from solvent-extraction columns. This solution, which carried a fraction of a gram per liter of plutonium residues, was then directed to a secondary recovery process (similar to the raffinate-treatment process of the Los Alamos accident). About 46 liters of solution containing 1400 to 1500 g of plutonium found its way into the transfer tank and led to the excursion. Apparently, most of the material was aqueous solution sucked up from a sump (into which it had overflowed from a geometrically favorable vessel) through a temporary line that had been used for cleanup. The total yield of 8.2×10^{17} fissions was distributed over 37 h with about 20% appearing in the first 1/2 h. Reconstruction of events indicated that an initial spike of about 10^{16} fissions was followed by smaller spikes throughout a period of 20 min, after which boiling occurred. The excursion was finally stopped by the boiling off of about 6 liters of water and the settling of some organic matter after it had extracted plutonium from the aqueous phase.

The initial burst (accompanied by a blue flash) triggered radiation alarms, and the plant was evacuated promptly. One man who was 5 or 6 ft from the transfer tank received a radiation dose of 110 rem, another perhaps 9 ft away received 43 rem, and a third at 26 ft received 19 rem. A unique feature of the postevacuation analysis of events was the use of a small, remotely controlled robot equipped with

television. This device, normally used for handling irradiated fuel, was used to fix the location of the incident, place meters and read them, and operate valves.

There were already plans to replace Recuplex, and the old plant was not reactivated after the accident. The modern plant makes fuller use of geometrically favorable equipment and vessels containing neutron poisons, it is adaptable to a variety of uses without improvisation, and its new equipment is easier to keep clean. It is recognized that the required flexibility of a salvage plant calls for special effort to maintain up-to-date written procedures that represent realistic practice.

The United Nuclear Corporation, Wood River Junction, R.I.—July 24, 1964. The scrap-plant facilities of the United Nuclear Corporation were designed to recover enriched uranium from scrap associated with the fabrication of reactor fuel. Initially, pickle liquor from fuel cleaning was being processed. Operations, which had started in March, were still preliminary when the accident occurred. The solution treatment, which involved geometrically safe primary equipment, consisted of normal solvent-extraction operations, trichloroethane wash of the resulting aqueous solution, solvent recovery with sodium-carbonate solution, concentration of the uranium solution by means of an evaporator, and precipitation as ammonium diuranate.

Because of startup difficulties, there was an unusual accumulation of contaminated trichloroethane, from which uranium was recovered by tedious hand agitation with sodium-carbonate solution. This led to improvisation of an easier process, in which the trichloroethane was treated in a tank intended only for makeup of the sodium-carbonate solution used in the normal recovery process. This tank, of nonsafe geometry, was the site of the excursion. Neither the plant superintendent nor one shift supervisor (of three) was aware of this practice. Meanwhile, solutions of unusually high ^{235}U concentration resulting from cleanout of a plugged portion of the evaporator had been stored in the same kind of 5-in.-diam bottles that contained the contaminated trichloroethane. Apparently, a bottle of the concentrated solution was mistaken for trichloroethane and poured into the 18-in.-diam sodium-carbonate makeup tank.

According to the most plausible reconstruction of events, two excursions occurred about 2 h apart. The first, a single spike of $\sim 1 \times 10^{17}$ fissions, took place when most of the concentrated solution had been poured into the tank. The shock splashed about one-fifth of the solution out of the tank and knocked the operator onto the floor. A flash of light was observed. The victim, who ran out of the building, had received an exposure estimated to be 10,000 rad, and died 49 h later.

It appears that enough solution was lost (final content 41 to 42 liters with 2 kg of uranium mostly as precipitate) so that the vortex from a stirrer in the tank was sufficient to maintain a subcritical state. Two h after the first excursion, however, two men re-entered the area and turned the stirrer off and then on again some minutes later, after which they drained the tank. (The radiation alarm was still sounding as a result of the original burst.) Apparently the second excursion occurred shortly after the stirrer was turned off. It could have been either a single burst or a sequence of bursts; the total yield of the two excursions was 1.3×10^{17} fissions. The two who drained the tank received radiation doses of 60 to 100 rad, and exposures of others who had been in the plant were minor. (They were 40 ft or more from the first excursion.)

After the accident, the United Nuclear Corporation took action to analyze methods of operation, including penetrating reviews of operating procedures, criticality limits and controls, uranium accountability and material balance, health-physics procedures and controls, training, and emergency procedures. Geometrically safe equipment for recovering uranium from trichloroethane was put into operation. (An alternative could have involved circulation through a vessel packed with raschig rings.)

Observations. In one sense, the accident experience in the nuclear industry has been too good. Six accidental excursions in 20 yr of processing fissile material are obviously insufficient to give a comprehensive picture of the ways in which criticality can occur and of the range of consequences. So we must be cautious about generalizing observations, including our introductory listing of common features of the accidents.

It is not surprising that all incidents have occurred in recovery plants, for the variety of

materials to be processed there requires flexibility that is not inherent in regular production operations. Furthermore, some of the plants involved were built in the early days when there had to be more reliance on the control of batch size than is typical of modern facilities. It is somewhat surprising, however, that all the excursions involved simple solutions instead of scrap dissolvers, because sampling difficulties made the old batch control of charges for dissolvers particularly unreliable. Although the absence of a dissolver accident in the older plants is partially attributable to large "ignorance factors" combined with normal "safety factors," there also seems to have been a measure of good luck. Now, improved criticality information makes it possible both to reduce the "ignorance factors" and to decrease the former dependence upon batch-size control. We will emphasize methods of improving safety under these conditions in later sections.

The observed range of excursion characteristics, lack of damage, and absence of public hazard are considered typical of solution accidents, although disruptive pressures and consequent public exposure are possible in unusual circumstances. Certain types of accidents with solid fissile material, particularly with ^{235}U metal, are more likely to be violent.^{1,3} Fortunately, it is not difficult to foresee the conditions, such as the falling together of large pieces of metal, that might lead to an extreme accident. Control of these conditions is usually straightforward and is emphasized in plant operations. Properties of solution excursions are illustrated further by an extensive series of kinetic experiments conducted at the Dijon Laboratory of the French Commissariat a l'Energie Atomique.⁴

As suggested by our review, only the radiation from an excursion is a sufficiently reliable characteristic for identification. Advance warning cannot be expected unless conditions are most unusual. Thus, in the absence of shielding, exposure is determined by the number of fissions and the distance from the excursion. This is shown by the crude correlation of Fig. 1, which is derived from observed exposures adjusted to a yield of 10^{17} fissions. In most instances of multiple bursts, evacuation eliminated or reduced exposure from all but the initial burst. The one positive conclusion of our review is that human

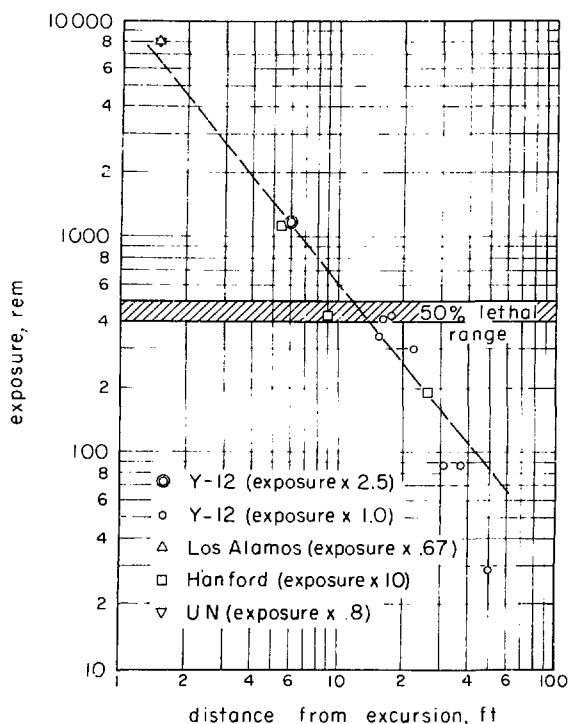


Fig. 1. Approximate correlation of exposure with distance from a solution excursion of 10^{17} fissions.

life has been saved by radiation alarms coupled with effective evacuation procedures.

This introduction to the subject of criticality control has been somewhat haphazard, for that is the way of history. Consequently, a summary may be in order. Pitfalls of the administrative control of batches and concentrations have been illustrated, particularly the problem of maintaining effective up-to-date procedures for flexible operations. Concepts of control by "favorable geometry" and by introduction of "neutron poisons" have appeared in discussions of corrective action. These ideas, of course, will require much amplification before they take on real meaning. Note that specific corrective measures appear to be straightforward once the causes of an accident become clear. This supports the thesis that an important requirement for nuclear safety is familiarity with all aspects of a process and the consequent ability to predict what can go wrong, which requires the talent of the process engineer.

A common feature of considerable significance is that each accident has been the result of a chain

of events, no one of which would have been harmful by itself. This suggests the usefulness of supplementing direct methods of control by small, generally inexpensive, precautions that tend to break chains of undesirable events. Generally, these aids are reminders, arrangements of material, and layout of equipment that encourage proper operations and make improper operations unnatural. It is considered wise to take advantage of a free precaution although it may appear unnecessary. The few opportunities to illustrate this type of aid will not be comprehensive, because full exploitation depends upon ingenious adaptation of the detail within a plant.

CRITICALITY RISK IN PERSPECTIVE

How criticality risk compares with risks from more conventional hazards is shown in *Operational Accidents and Radiation Exposure Experience Within the United States Atomic Energy Commission, 1943-1970* (USAEC Division of Operational Safety, Washington, D. C., 1965). Although licensed operations are not included, the experience of AEC contractors gives a good picture because of their lengthy work record and pertinent activities. We show the character of fatalities attributable to plant and laboratory operations in Table I. Plant criticality, with its single death (the other death was not in an AEC installation), ranks with gunshot and drowning, not with the more common hazards such as electric shock, explosion, burns, and falls or falling objects.

TABLE I
FATALITIES IN AEC PLANT AND LABORATORY
OPERATIONS, 1943-1970
(Construction and Direct Government
Activities Excluded)

| Accident Category | Fatalities |
|----------------------------|------------|
| Plant criticality exposure | 1 |
| Critical assembly exposure | 2 |
| Electric shock | 18 |
| Motor vehicle, aircraft | 21 |
| Chemical explosion | 12 |
| Burns | 12 |
| Falls, falling objects | 14 |
| Asphyxiation, suffocation | 9 |
| Poison | 3 |
| Reactor explosion | 3 |
| Drowning | 2 |
| Gunshot | 1 |
| Total | 98 |

Note that inclusion of AEC construction and direct government activities would increase the total number of fatalities from 98 to 295. In terms of the other common safety indexes, radiation accidents of all kinds accounted for 0.4% of injuries and 2% of time lost.

Although this favorable record speaks well for the methods of criticality control, we do not wish to suggest that it is reason for relaxation. To maintain a good record, improved control techniques, especially those designed into processes, must, presumably, keep up with the foreseeable greatly increased demands for fissile material. In the past, improvements have equaled production increases, and there is reason to believe that this neck-and-neck process can continue.

DEFINITION OF NUCLEAR CRITICALITY SAFETY

Nuclear criticality safety is usually defined as the art of avoiding a nuclear excursion, and, indeed, this is the usually practical viewpoint. However, we should recognize the situation demonstrated by the Idaho incident of January 1961, in which the consequences of an excursion were trivial. A process may be designed to include shielding, confinement, and other conditions like those at Idaho so that the probability of an excursion may be allowed to increase. In at least two instances, this alternative has proved less expensive than an unshielded process with the appropriate added restrictions.

Perhaps, then, nuclear criticality safety may be defined more precisely as protection against the consequences of a nuclear excursion. Although this extended definition points out a flaw in our use of "criticality control" as a synonym for "nuclear criticality safety," we shall continue to treat these terms, and simply "nuclear safety," as equivalent.

PRACTICAL NUCLEAR SAFETY FUNDAMENTALS

One purpose of this section is to lay the groundwork for a practical "philosophy" developed throughout the rest of the report. As explained further in Sec. V, this philosophy is not specific to criticality safety, but is based upon safety principles that were developed and tested before fissile material appeared on the scene. Points of view that we have attempted to introduce for this reason may be stated more specifically as follows.

1. Safety is an acceptable balance of risk against benefit; it is meaningless as a concept

isolated from other goals. It follows that safety should be considered one of the goals of design and operation instead of something superposed.

Although experience has shown that criticality hazards are no more serious than other industrial hazards, controls for balancing criticality risk against benefit are somewhat more stringent than is usual in nonnuclear industry. It is reasonable that there be some allowance for the uneasiness naturally associated with this new type of hazard. But the extreme concept of risk elimination (as implied by any claim that certain controls "assure" safety or "ensure" safety) is dangerously misleading. Dismissing risk as nonexistent can detract from the continuing job of maintaining an acceptably low risk level.

2. Accident prevention depends upon responsibility for safety implementation (and commensurate authority) at the supervisory level closest to operation, under the general direction and policies set by higher management. Attempts to control detail at a remote level are misguided.

Because of the requirement for governmental regulation, great care is required to preserve this precept in criticality safety. Remotely administered detail discourages the on-the-job alertness required for effective control, because it encourages the attitude, "Someone else is taking care of us."

3. Safety regulation should be based upon professionally generated standards and should preserve alternative routes to safety objectives. The arbitrary selection of a single route (as by rule) may eliminate the best economic balance or the most convenient scheme.

Inflexible rules hamstring the designer in his traditional search for the most satisfactory way to fulfill his many objectives. The result is to set safety apart from other objectives, and increase the chance of an awkward operation that invites improvisation. Flexibility frees the designer to apply to integrated process design the considerable experience that has accumulated in nuclear industry.

4. Other things being equal, simple, convenient safety provisions are more effective than complex or awkward arrangements. Similarly, "free" (no cost) contributions to safety should be nurtured.

As an example of this principle, criticality safety is enhanced by arrangements of material and equipment that tend to make proper operations convenient and maloperation inconvenient.

These principles of nuclear safety will be interpreted further in Sec. V, where the intent is to suggest reasonable ways to compare conditions that may normally be encountered in an operation to corresponding critical conditions. To prepare for this, however, we shall consider in Secs. II-IV the scope of existing information about critical configurations.

II. FUNDAMENTALS OF CRITICALITY

QUALITATIVE CRITICALITY CONCEPTS

By the statement that a configuration of fissile material is just critical,* we mean that the average power from fission is constant. In this condition, one of the several neutrons from the typical fission process produces a new fission. The remaining neutrons are either lost by capture (non-fission absorption) or by escape from the system (leakage). Thus, the constant fission chain reaction is linked by neutrons that are held in delicate balance by just the right competition between fission and capture plus leakage. The features of fissile systems which can influence criticality are most easily pictured in terms of these competing fates of neutrons.

To develop this picture of effects on criticality, we must recognize one more aspect of neutron behavior. The most likely occurrence when a typically high-energy neutron from fission strikes any nucleus is that the neutron will simply be deflected. (As we shall see, this is not the most probable process when a low-energy neutron strikes a fissile nucleus.) If the nucleus is heavy, the neutron loses little energy because of such a collision, but the lightest nucleus, hydrogen (about the same mass as the neutron), may cause great neutron-energy loss. The process of energy loss during successive collisions of a neutron with light nuclei, as in passing through water, is called "neutron moderation." The importance of moderation is that the chance of producing fission during a collision with a fissile nucleus increases greatly as the neutron energy becomes small.

*Strictly speaking, this is the "delayed critical" state.

Now we are ready to use the simple concepts of neutron behavior to develop intuition about criticality. Let us illustrate the various influences on criticality by limiting our attention to two common materials, enriched uranium and water. To start, we consider a critical sphere of U(93) metal at normal density. The diameter of this sphere is about 6.9 in., corresponding to a volume of 2.8 liters and a total mass of about 52 kg. If the same quantity of material is formed into a slab or an elongated cylinder, distances through which neutrons must scatter to reach a surface are decreased (the surface-to-volume ratio increases), so the chance that a neutron may escape from the material is increased. In other words, leakage is increased at the expense of fission and capture, so that the new shapes are subcritical. Returning to the sphere, if the size is maintained but the density of U(93) is decreased, neutrons pass through less matter on their way to the surface, the chance of leakage is increased, and the new sphere is subcritical. Likewise, a decrease in ^{235}U enrichment at constant size and density decreases the chance of fission relative to leakage and capture, so that the sphere is again subcritical.

Now, several different influences of water on our U(93) sphere will become apparent. If the sphere is immersed in water, some neutrons that would otherwise escape from the surface are scattered back into the fissile material, leakage is reduced, and the sphere is supercritical. Actually, the critical diameter of the uranium sphere drops to 5.3 in. (corresponding to 1.3 liters or ~ 24.5 kg of uranium). Of course, this neutron-return effect is by no means limited to water. Any material that surrounds the fissile sphere will act similarly as a neutron "reflector." Objects at a distance from the sphere will have reduced neutron-reflection effects, but even if these objects are fissile, they may be viewed as reflectors.

If, instead of surrounding the sphere, water is mixed homogeneously with the U(93), there are striking changes in neutron economy as the proportion of water increases. When the volume of water is not much greater than that of the uranium in the mixture, the water's moderating effect is not enough to offset the effect of reducing the uranium density. (Collisions with hydrogen are too few for significant reduction of neutron energy.) Consequently, the

quantity of uranium that was critical without dilution is now subcritical. With further water dilution, however, the effect of moderation takes over (there are more and more collisions of neutrons with hydrogen), and the system becomes increasingly supercritical. This trend continues until the volume of water is about 350 times that of uranium, beyond which neutron capture by hydrogen offsets any additional effect of moderation. (Although the chance of neutron capture during one collision with hydrogen is small, each neutron undergoes many collisions at this dilution.) Here the critical diameter of a bare sphere is roughly 15 in. and the volume is up to 30 liters, but the ^{235}U mass is only 1.4 kg. A complete water reflector around the sphere reduces these values to about 12-in. diam, 16 liters, and 0.8 kg of ^{235}U . These last conditions are of special significance in that they represent the minimum critical mass of ^{235}U encountered in usual processes.

Figure 2 pictures the complete influence on critical mass of the shifting competition among ^{235}U density, moderation, and hydrogen capture. The curve applies to bare spheres consisting of U(93) metal mixed with water. As the uranium becomes very dilute, the effect of hydrogen capture becomes progressively more important. Finally, at a water volume about 1600 times that of the uranium, this capture predominates over all other effects, and the critical mass becomes infinite. This limiting critical condition corresponds to a concentration of 11 g of ^{235}U /liter of aqueous solution.

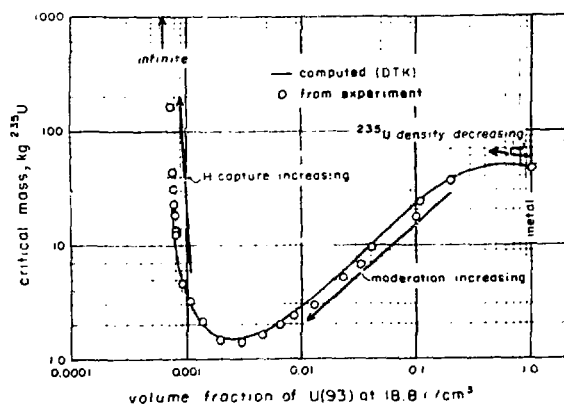


Fig. 2. Critical masses of bare spheres of homogeneous U(93)-water mixtures.

ORIGINS OF CRITICALITY DATA

So far, the few numbers we used were primarily to emphasize a qualitative picture of neutron behavior to help us "understand" criticality. Now, however, numbers become of central importance, because the next step toward "understanding" must be quantitative. At this point, a diversion into the sources of criticality data (our "numbers") will help to pave the way.

The development of information about criticality has been considered a responsibility of the reactor physicist or reactor engineer. Broadly, reactor-physics methods are either experimental or are computational substitutes for experiment. To qualify as a general substitute, the computational method must apply to a wide variety of compositions and must reproduce the effects of all neutronic processes that occur in real systems. Methods capable of such versatility,* although conceptually straightforward, are so complex numerically that they require the use of a high-speed, high-capacity, electronic computer.

Like experimental results, computed critical conditions must be evaluated for reliability before they can be accepted. Indexes of accuracy, such as probable error or standard deviation, are not directly available from calculation as they are from experiment, although exploration toward this end should be noted. Lacking such indexes, the only means of judging the reliability of a computational scheme is to compare its results with a wide selection of experimental data. In Fig. 2 for example, the points are derived from experiment, and the solid curve is computed by a technique known as the "DTK code with Hansen-Roach cross sections" (We will use DTK as an abbreviation). The probable error of the experimental points in Fig. 2 is within the size of the identifying symbol (except at a volume fraction of 0.1), and the reliability of the computed curve can be judged by its departure from the experimental points. In broader regions without experimental data, the solid line must be interpreted conservatively if it is to be used for nuclear-safety evaluation. This requires the help of the specialist and judgment

* These methods are Monte Carlo and multigroup diffusion and transport techniques with specified inputs. The DTK method (equivalent to S_n and DSN) is a multigroup transport code. The version most widely used as of this writing is DTF-IV, a slight modification written in FORTRAN.

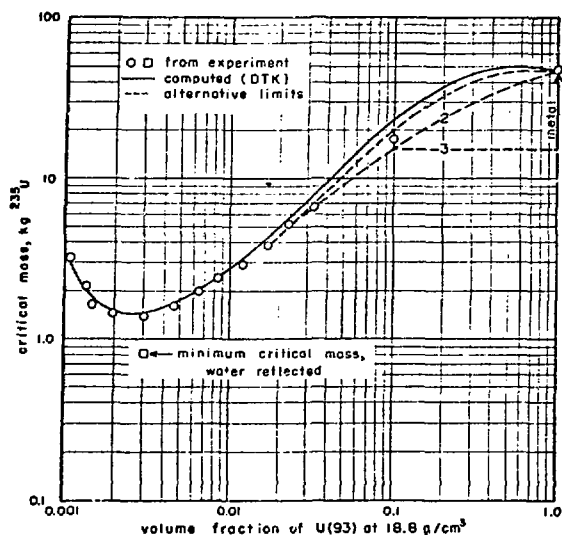


Fig. 3. Alternative safety limits for critical masses of bare spheres of U(93)-water mixtures.

based on his experience and insight. The reasonableness of his conclusions, which he should attempt to make apparent, can be decided in any of a number of ways.

For example, not long ago the experimental points for bare, water-moderated spheres appeared as in Fig. 3 instead of as in the more modern Fig. 2. Throughout a large region of higher uranium concentrations the computed curve (solid) had little experimental support. The three dotted curves show various ways in which the computed curve could then be scaled down for nuclear-safety application.* Curve 1, which carries a constant fractional shift throughout the unsupported region, was not considered conservative enough. Curve 3, at the other extreme, is too conservative because it levels off abnormally where critical mass is known to be increasing. Even without much insight, however, the more carefully selected curve 2 appears reasonable. At this point, it is worth noting the influence of the recent experimental points that appear in Fig. 2 but not in Fig. 3. It turns out that curve 1 of Fig. 3 is adequately conservative, although this should not

*Note that unreflected spheres are useful for nuclear-safety application only if effects of ever-present reflection are superposed. A more realistic example, which would involve water-reflected spheres, happens to be a less clear illustration.

have been assumed in advance. Thus, the difference between curve 1 and curve 2 represents an "ignorance factor" that was removed by the new experimental information.

This illustration is typical of a situation that will confront us time after time when experimental information is inadequate. In later discussions we will have to use computed data, both for illustration and for nuclear-safety application. Unless specifically stated otherwise, the DTK technique with Hansen-Roach cross sections will be represented, because its results have been compared abundantly with experiment.⁵

TYPICAL DETAILED COMPUTATIONAL TECHNIQUES

A credibility gap should be expected whenever a calculated number is simply handed from one person to another for criticality safety guidance. In addition to the question of inherent accuracy of the calculation, how does the user know that there has been no input error or machine error? Our first suggestion is that the user have some acquaintance with the capability of the computational technique.

DTK. As one example, let us consider the DTK code with Hansen-Roach cross sections.⁶⁻⁸ This "transport" code describes the probable behavior of a large population of neutrons within a system of elementary geometry. The description is in terms of an adaptation of the integral equation that Boltzmann developed for problems of gas dynamics which are similar to neutron-transport problems but even more complex. The simplified Boltzmann equation is integrated numerically, giving relative values for gain of neutrons by fission and for loss of neutrons by capture and leakage to a precision that is limited only by machine capacity and available running time. A principal option of DTK adjusts dimensions to balance neutron gain and loss, thus establishing critical size.*

As contrasted with precision, the accuracy of a DTK result is controlled by the cross-section set that is part of the problem input (the rest of the input describes the geometry and composition). Neutron cross sections of an isotope give the probabilities of various neutron interactions with that isotope, namely, capture, fission, and scattering (with

*Another common option establishes the neutron reproduction number, k , for a system of fixed size.

or without energy loss). They depend upon neutron energy, and for DTK are given as average values for each of a limited number of neutron-energy subdivisions or "groups."

The Hansen-Roach cross-section set has 16 of these neutron-energy groups. The virtue of this relatively old, but stable, set for safety purposes is that it has been used to check almost all appropriate experimental critical masses.^{5,9,10} Thus biases of results are known for critical systems with a wide range of composition, and they can be estimated for most new compositions. In brief, calculated and experimental critical masses agree to better than 10% (a value smaller than usual safety margins) for spheres of uranium at enrichments of about 5% ²³⁵U or greater in combination with many other materials and for hydrogen-moderated plutonium or ²³³U, either bare or water-reflected. Nonconservative biases (calculated values too large) occur for hydrogen-moderated uranium with enrichments below about 5%. Discrepancies of about 15% are found with 2% enriched uranium, and results deteriorate further at still lower enrichments, showing about 30% discrepancy for U(1.4). Although improved agreement would be expected of an up-to-date cross-section set, the burden of reestablishing biases has prevented modernization of the generally adequate Hansen-Roach set.

A limitation of DTK is that it handles only the one-dimensional geometries, spheres, infinitely long cylinders, and slabs of infinite extent. In addition to individual units of these shapes, a "cell" option of the code covers infinite lattices that can be approximated by close-packed spheres or by close-packed cylinders of infinite length. Practically, the geometric limitation of DTK is not so severe as it might seem, because results may either lead directly to conservative safety limits for other forms that fit into the calculated geometric envelope, or, as we shall see in the next section, be converted empirically to apply to shapes such as finite cylinders and parallelepipeds.

The essential simplicity of one-dimensional forms is that the average behavior of neutrons depends upon only the neutron energy, the materials encountered, and a value of angle at which each surface is crossed. (Location on the surface need not be considered because all such points for a sphere, infinite cylinder, or infinite slab are equivalent.)

For other forms, behavior depends upon the location of the surface crossing as well as its angle, which greatly complicates numerical solution. Although two- and three-dimensional transport codes have been developed, they either require long computing time on machines with large storage capacity, or involve relatively crude geometric and cross-section approximations. For criticality-safety applications, such codes do not generally compete with the Monte Carlo techniques that we will consider next.

Monte Carlo.¹¹ Instead of treating the probable behavior of a large neutron population directly as do transport codes, the Monte Carlo technique mathematically traces the paths of individual neutrons, collision by collision, until their disappearance because of processes such as absorption and leakage. Changes of energy, direction, and position from one collision to the next are governed by probability distributions that are derived from tables of cross sections vs neutron energy. These distributions are sampled statistically to establish the successive steps in each path. The result of superposing many such neutron histories simulates an extremely low-power critical experiment, even including the meanderings of neutrons throughout the assembly.

An advantage of Monte Carlo over transport methods is that it is inherently less limited by geometry; the computing effort required to track neutron paths is relatively insensitive to geometric complexity. As a result, Monte Carlo can handle any geometry, generally as an assemblage of cells—for example, finite cylinders and hemispheres, or combinations thereof, and readily accommodates finite arrays of units.

As must be suspected, straightforward Monte Carlo simulation would usually require impossibly great machine capacity and computing times. But clever sampling tricks and weighting techniques to emphasize the most important interactions have made the method practical for a large variety of systems, though it is still a more expensive type of calculation than DTK. For criticality calculation, Monte Carlo efficiency decreases with increasing moderation because of the many scattering steps required for significant reductions of neutron energy.

Most Monte Carlo codes are equipped to use detailed cross-section information instead of neutron-energy-group averages. One Oak Ridge version known

as KENO,¹² however, uses the DTK multigroup approach with Hansen-Roach cross sections, largely to save computing time when more detailed treatment is not required. Many checks of experimental data by this code confirm the biases established by DTK and indicate that geometric complications do not influence them.¹³ Assuming adequate precision, which depends only upon the number of neutron histories considered, it thus seems that the accuracy of KENO (as of DTK) is governed by the cross-section set, not by the numerical manipulations.

This brief discussion of representative transport and Monte Carlo criticality codes, and of the Hansen-Roach cross-section set, is to remove some of the mystery associated with mere names and to give some feeling for their capabilities and limitations. We conclude that either DTK or KENO can provide results as good as the inherent accuracy of the cross sections used, and that properly computed results can be applied reliably when (as is usual) biases introduced by the cross-section set can be estimated. DTK is the cheaper and more convenient method whenever the needed information can be deduced from one-dimensional results. KENO is appropriate for complex geometry or arrays, but becomes expensive for well-moderated systems. Other codes and cross-section sets can be examined similarly, but may require special checks against experimental data to confirm their applicability.

The Acceptance of Computed Data. Knowing the capability of a computational technique is only a start toward accepting a result as reliable. The most common causes of failure to achieve that capability are input errors (cross sections, material designations, or dimensions may be transcribed improperly), or misapplication of techniques (to be illustrated later). Self-monitoring features of modern machines usually inform the operator if machine errors occur.

Broadly speaking, the supplier of calculated data should be able and willing to support the validity of the numbers he provides—to tell how he was convinced that they are reliable. The user, of course, should be a good enough judge of the supporting arguments to understand their implications and to guard against a "snow job." The ways in which confidence can be established depend upon the experience and ingenuity of both the supplier and user of the data, upon similarity of results to experimental

data, and upon the nature of information in printout sheets provided by the computing machine. There are few real rules; rather there are primarily suggestions, and illustrations based on calculations with the DTK code and Hansen-Roach cross sections.

The printout, with which the supplier must be familiar, reproduces input data (geometry, materials, and cross sections) and the spatial and angular subdivisions used for numerical integrations, gives computed neutron spectra for the various spatial regions, and indicates formal completion of the problem (the degree to which convergence criteria are satisfied). Thus, a simple but careful check of the reproduced input data can eliminate input errors (which are not uncommon). This reassuring check need not be repeated for parts of input that carry automatically through succeeding problems.

Other steps toward establishing confidence are less specific. A simple-minded observation is that results should look reasonable in terms of background criticality data. As was not true earlier, there is now usually a basis for a fair guess of what the answer should be. Even better than judging the plausibility of a single result is the added possibility of examining trends from a series of related calculations. For example, it is sometimes easier to estimate relative effects of simple compositional changes than to estimate critical sizes. Therefore, multiple calculations are often considered worthwhile when only a single criticality value is actually required.

Still other clues about the validity of a calculation are given by the spectra that appear on the printout—whether mean energy and shape are about right, and whether spectral changes from one medium to another (such as the energy decrease expected when passing from a metal core into a hydrogenous reflector) make sense. Spectral information may be especially useful when the calculated critical size is suspect, for it may point out an undetected error.

The other main source of potential error is a poor choice of spatial and angular subdivisions for the numerical integration performed by the machine. Although anyone experienced in criticality calculation will avoid such a difficulty almost automatically, the user still has the right to be convinced that his problem was not set up too crudely. Explanation may serve, but the most satisfying demonstration is

a check of experimental data against similar calculations. Where use of a criticality code is standardized, a backlog of experimental checks may provide the desired reassurance.

After establishing that the calculation is in order, one must decide what allowance to make for bias introduced by the cross-section set. As we have noted, a 10% reduction of critical sphere mass computed using the Hansen-Roach set would cover biases for most compositions except moderated uranium at enrichments below about 5% ^{235}U . This translates to no worse than a 10% reduction of infinite-slab thickness or cross-sectional area of an infinite cylinder. Naturally, the judgment required in applying established biases to a computed system depends upon the extent to which components of the new system are represented by families of experimental assemblies. For example, a bias within 10% might be inferred for a plutonium-iron sphere reflected by natural uranium, because that bias is not exceeded by spheres of water-reflected U(93), water-reflected plutonium, natural-uranium-reflected U(93), and bare U(93)-iron near the proportion of interest.

Frequently an arbitrary 20 to 30% reduction of computed critical mass (before introducing the safety margin) can be introduced without compromising the plant process. Such an allowance, of course, greatly reduces the need for detailed evaluation.

SIMPLE COMPUTATION

Now, what of the simplified computational methods* that are emphasized in reactor-physics books? As indicated before, we do not consider them a valid source of data for criticality control. When used to interpret experimental results, however, they can sharpen the picture of neutron processes that influence criticality. Although they may thus contribute to "understanding," we simply refer those who desire this extra capability to the excellent reactor physics texts available.¹⁴⁻¹⁶ Our approach to criticality will continue to be empirical, with interpretation depending upon the qualitative picture of neutron behavior.

Another contribution of simplified reactor theory is to suggest forms for certain empirical correlations. One of the more useful of these is a relationship that permits us to deduce criticality data

*These methods include the four-factor formula, age theory, and one- or two-group diffusion theory.

for any simple configuration from the data for a single shape such as a sphere. In the next section, we shall accept the logic of the form and proceed to examine this geometric relationship as an operational tool.

Before concluding this brief discussion of reactor theory, we should mention two common indexes of criticality. The first, the reproduction number, k , is the ratio of the average rate of neutron production to the average rate of loss (by absorption and leakage). Naturally, a fissile system is just critical if $k = 1$, subcritical if $k < 1$, and supercritical if $k > 1$. The reproduction number is a favorite of theoreticians and requires theoretical help for interpretation if it differs from unity.

The other index, called "buckling" and symbolized by B^2 , depends only upon the composition of the fissile system and is a measure of the critical size. If buckling is negative, the fissile composition is subcritical at any size; if zero, the composition is critical only at infinite size. Positive values correspond to finite critical sizes. Elementary theory gives expressions that relate B^2 to critical dimensions of systems of various shapes. These are the expressions that provide the form of empirical relations for converting from one critical shape to another.

III. CRITICALITY INFORMATION—INDIVIDUAL UNITS AT NORMAL DENSITY

The purpose of this section and the next is to give quantitative significance to our understanding of the various factors that influence criticality.

*The story of the name "buckling" is given by Weinberg and Wigner on p. 203 of their book, *The Physical Theory of Neutron Chain Reactors*.¹⁵ Briefly, neutron-diffusion theory says that flux distributions, $\phi(x)$, throughout cores of fissile material satisfy the wave equation $\Delta\phi(x) + B^2\phi(x) = 0$, where $\Delta\phi(x)$ is the Laplacian of $\phi(x)$. From this expression, $B^2 = \Delta\phi(x)/\phi(x)$, which means that B^2 is a measure of the curvature (or warping) of the $\phi(x)$ distribution. This observation suggested the name "buckling."

The value of B^2 is related to critical core dimensions as a consequence of a requirement that the flux extrapolate to zero at a certain distance beyond the surface (the extrapolation distance). The resulting relationships for cores of several shapes appear as Eqs. (1a) to (1f) of our text. Shape-conversion relationships such as Eq. (2) which are the result of assuming the same value of B^2 for cores of different shapes, define conditions of equal neutron leakage.

The emphasis is on data that contribute to this objective as opposed to comprehensive coverage of criticality information. For a reasonably complete collection of experimental critical-mass data, see AEC report TID-7028, "Critical Dimensions of Systems Containing U^{235} , Pu^{239} , and U^{233} ."¹⁷ Although we assume that TID-7028 will be generally available, we have reproduced some of its more essential figures as our Appendix.

Because of the gaps that exist in experimental data, results of calculation must be used for many of our illustrations. As mentioned before, these computed data should be evaluated for reliability before being used for nuclear-safety purposes. In many cases, existing compilations of computed vs experimental critical sizes will be adequate for such an evaluation.⁵

In the course of illustrating influences on criticality, we shall discuss several generally useful empirical relationships. Here, more than usual detail will be included in the expectation that these relations can become practical tools of all who are concerned with criticality control. The first of these empirical tools is the scheme for shape conversion.

RELATIONSHIPS FOR SHAPE CONVERSION

Expressions that derive from simple reactor theory relate buckling, B^2 , and the dimensions of critical cores of fissile material of easily described shapes.¹⁴⁻¹⁶ As in each case that follows, the actual dimension is augmented by a so-called extrapolation distance, δ , which we will discuss further.

For a sphere of critical radius r_s ,

$$B^2 = \left(\frac{\pi}{r_s + \delta} \right)^2. \quad (1a)$$

For a parallelepiped of critical dimensions h_1 , h_2 , h_3 ,

$$B^2 = \left(\frac{\pi}{h_1 + 2\delta} \right)^2 + \left(\frac{\pi}{h_2 + 2\delta} \right)^2 + \left(\frac{\pi}{h_3 + 2\delta} \right)^2, \quad (1b)$$

from which the expression for a critical cube of side h_1 is

$$B^2 = 3 \left(\frac{\pi}{h_1 + 2\delta} \right)^2. \quad (1c)$$

For a cylinder of critical radius r_c and critical height h_c ,

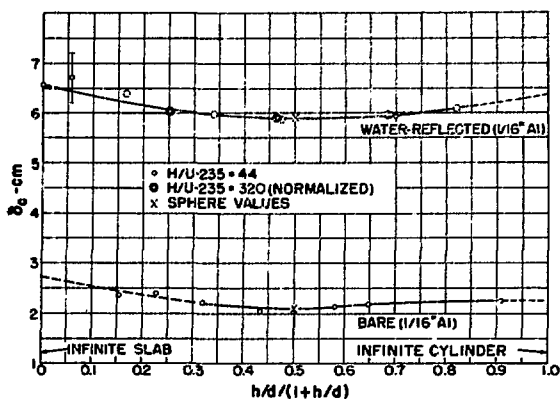


Fig. 4. Extrapolation distances for shape conversion of U(93) solutions. The curves apply to cylinders of height h and diameter d .

$$B^2 = \left(\frac{2.405}{r_c + \delta} \right)^2 + \left(\frac{\pi}{h_c + 2\delta} \right)^2. \quad (1d)$$

In the special case of an infinitely long cylinder of critical radius r_c ,

$$B^2 = \left(\frac{2.405}{r_c + \delta} \right)^2. \quad (1e)$$

and for an infinite slab of critical thickness h_c (a cylinder of infinite radius), the buckling becomes

$$B^2 = \left(\frac{\pi}{h_c + 2\delta} \right)^2. \quad (1f)$$

For any given fissile composition, we recall that B^2 is a constant, so these various geometric expressions for B^2 can be equated. And, according to simple theory, the "extrapolation distance," δ , is a constant for each type of reflector that surrounds the fissile material. Actually, experiments that have compared various critical shapes of a fixed composition show that the quantity δ is not quite constant, but depends somewhat upon the shape. The way in which δ varies with the elongation of critical cylinders is shown by Figs. 4 and 5, taken from TID-7028.¹⁷ The shape of each cylinder is characterized by the ratio of its height to its diameter (h/d). To avoid infinitely long curves, however, δ appears in the figures as functions of $(h/d)/(1 + h/d)$ so that the abscissa zero corresponds to a slab of infinite diameter, and unity corresponds to a cylinder of infinite length.

Figure 4 applies to U(93) solutions, the upper curve to water-reflected cylinders, and the lower to essentially bare cylinders. Data for more limited.

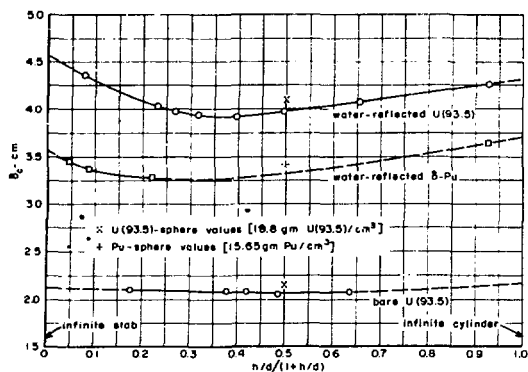


Fig. 5. Extrapolation distances for shape conversion of U(93.5) and plutonium metal. The curves apply to cylinders of height h and diameter d .

families of cylinders suggest that the lower curve of Fig. 4 also applies to ^{233}U and ^{239}Pu solutions (with concentrations similar to those indicated for ^{235}U), and that the upper curve may be used for ^{233}U solutions if scaled by the factor 5.1/5.9, and for ^{239}Pu solutions if scaled by the factor 5.7/5.9.

The other experimentally established values of extrapolation distance apply to U(93) metal and plutonium metal, and appear in Fig. 5. This leaves uncertainties about appropriate values of δ for slurries and comp compounds that are intermediate between the dry metal and solutions, about very dilute solutions, and about any homogeneous mixture of uranium at reduced ^{235}U enrichment. Here, it is necessary to resort to results of conservative calculations until further experimental data become available. Sometimes two components of the extrapolation distance for a reflected system may be estimated separately. A value δ_c , from the upper curve of Fig. 4, for example, may be viewed as consisting of the extrapolation distance for the bare cylinder of the same shape, δ_o (from the lower curve), plus a quantity, δ_r , that depends only upon the reflector and is called "reflector saving."*. If $\delta_c = \delta_o + \delta_r$ is desired, δ_o may be known and δ_r computed, or vice

versa. Values of reflector saving that apply to hydrogenous mixtures of uranium at enrichments of less than 93% ^{235}U may be derived, for example, from Fig. A1 in the Appendix. The reflector saving of water is simply the difference between critical radii of corresponding unreflected and water-reflected spheres.

Of course, the value of the ability to convert from one critical shape to another is that critical data for a single, simple shape such as a sphere may be applied generally. Thus the sphere, which appears so seldom in process equipment, actually represents other more practical shapes. (As an alternative to actual shape conversion, critical masses and volumes of spheres, which are minimum values, may be applied conservatively to other shapes.)

Some examples will clarify shape conversion by means of Relations (1) and empirical values of extrapolation distance. Suppose that we wish to see how the critical diameter of elongated cylinders changes with h/d for a water-reflected solution containing 500 g of U(93)/liter. At this concentration, chosen because it represents the minimum critical size for solutions, the radius of a critical water-reflected sphere is known to be 11.5 cm. Combining Eqs. (1a) and (1d), we have

$$\left(\frac{\pi}{r_s + \delta_s}\right)^2 = \left(\frac{2.405}{r_c + \delta_c}\right)^2 + \left(\frac{\pi}{h_c + 2\delta_c}\right)^2, \quad (2)$$

where the dimensions (including δ) on the left apply to a critical sphere, and those on the right apply to a corresponding critical cylinder for which $h/d = h_c/2r_c$. With $r_s = 11.5$ cm and $\delta_s = 5.9$ cm (from Fig. 4), Eq. (2) becomes

$$\left(\frac{2.405}{r_c + \delta_c}\right)^2 + \left(\frac{\pi}{2r_c(h/d) + 2\delta_c}\right)^2 = 0.0325. \quad (3)$$

For the infinitely long cylinder ($h/d = \infty$), Fig. 4 gives $\delta_c = 6.35$ cm, and the value $r_c = 7.0$ cm satisfies the above relation. Continuing with finite values of h/d , Eq. (3) may be solved by trial to give the results shown in Table II for various elongated cylinders.

* In some literature, terminology is confused by use of the term "reflector saving" to mean δ_c instead of δ_r .

TABLE II

CRITICAL ELONGATED WATER-REFLECTED CYLINDERS OF
U(93)O₂F₂ SOLUTIONS AT 500 g OF ²³⁵U/LITER

| h/d | δ_c (cm) | r_c (cm) | d(in.) | Crit. volume (liters) |
|----------|-----------------|------------|--------|--------------------------|
| ∞ | 6.35 | 7.0 | 5.5 | ∞ |
| 10 | 6.2 | 7.2 | 5.7 | 23.6 |
| 5 | 6.1 | 7.5 | 5.9 | 13.2 |
| 3 | 6.0 | 7.9 | 6.2 | 9.4 |
| 2 | 5.95 | 8.5 | 6.7 | 7.7 |

sphere: 6.4

Two features of this listing are worth pointing out. First, the value of the critical diameter of the infinitely long cylinder is not overly conservative when assumed for cylinders with height-diameter ratios as small as 5. This observation is useful where interest is in a long cylinder for which no experimental critical data exist, so that results of computation must be relied upon. The value for an infinite cylinder is much easier to compute than that for a finite cylinder, and, as just shown, it is almost as good. The second feature is that there is not much advantage over using the sphere critical volume until the cylinder height becomes several times its diameter. (The volume of the critical equilateral cylinder is about the same as that of the sphere.)

In the particular case of the infinite cylinder, the unreflected critical radius is obtained simply by adding the reflector saving to the water-reflected critical radius. For the first item of Table II, the reflector saving is $\delta_c(\text{refl}) - \delta_c(\text{bare}) = 6.35 - 2.25$ (from Fig. 4); so the critical radius of the bare infinite cylinder is $7.0 + 4.1 = 11.1$ cm, and the critical diameter is 8.7 in. Again, we remind ourselves that there is always some reflection, so the bare dimension is of little practical value except as a reference point. There are situations, for example, in which reflection is small enough that an average of the bare and reflected diameters would be appropriate for criticality control. There can be other instances in which the critical diameter of a long, bare cylinder is established experimentally and the reflector saving of water can be estimated. Then the critical radius of the water-reflected cylinder will be approximated by subtracting the reflector saving from the bare radius.

TABLE III

CRITICAL SQUAT WATER-REFLECTED CYLINDERS OF
U(93)O₂F₂ SOLUTIONS AT 500 g OF ²³⁵U/LITER

| h/d | δ_c (cm) | h_c (cm) | h_c (in.) | Crit. volume (liters) |
|------|-----------------|------------|-------------|--------------------------|
| 0 | 6.6 | 4.3 | 1.7 | ∞ |
| 0.05 | 6.45 | 5.0 | 2.0 | 39.5 |
| 0.1 | 6.35 | 6.0 | 2.4 | 17.0 |
| 0.3 | 6.1 | 9.6 | 3.8 | 7.8 |
| 0.5 | 5.95 | 12.8 | 5.0 | 6.6 |

sphere: 6.4

Interest in critical squat cylinders (or slabs) of solution is stimulated largely by the use of shallow pans to catch material that may leak from equipment. Here the critical depth of solution is important, so, to continue Table II for small height-diameter ratios, it is convenient to rewrite Eq. (3) as

$$\left[\frac{2.405}{\left(\frac{h_c}{2} / \frac{h}{d} \right) + \delta_c} \right]^2 + \left(\frac{\pi}{h_c + 2\delta_c} \right)^2 = 0.0325 \quad (4)$$

In this case, the simple solution of this equation is for a slab of infinite diameter ($h/d = 0$): Figure 4 gives $\delta_c = 6.6$ cm and $h_c = 4.3$ cm or 1.7 in. Again, the above relation must be solved by trial for finite diameters. Typical results are shown in Table III.

In this case, too, the critical volume does not increase significantly until the diameter becomes several times the height. Now, the thickness of the bare infinite slab is obtained by adding twice the reflector saving from Fig. 4 to the water-reflected value (to remove the effect of water on each side of the slab). The result, about 4.7 in., is again largely of academic interest. On the other hand, a semireflected slab simulates the usual catch pan that is reflected only on its base. The appropriate critical thickness, obtained by adding only a single-reflector saving to the value for full reflection, is then 3.2 in.

As mentioned, Tables II and III apply to a concentration at which solution critical dimensions are minimum, so they may be used as conservative estimates for other concentrations. Furthermore, critical parameters of the uranyl-fluoride solution are slightly smaller than those of the nitrate or other

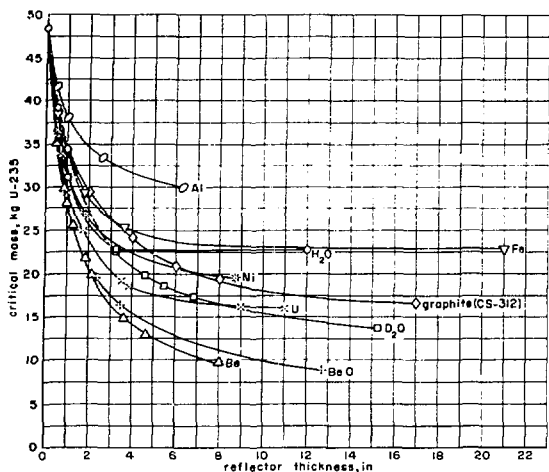


Fig. 6. Critical masses of U(93.5) metal spheres in various reflectors; $\rho(U) = 18.8 \text{ g/cm}^3$.

common aqueous solutions. Thus the dimensions in these tables provide a generally useful basis for evaluating the nuclear safety of ^{235}U solutions in isolated containers.

EFFECTS OF VARIOUS REFLECTORS

Water, the neutron reflector discussed so far, happens to be representative of reflection effects usually encountered in a processing plant. Figure 6 (taken from TID-7028) shows the most complete experimental set of comparisons of different reflector materials. The curves apply to various thicknesses of reflector surrounding spherical U(93) metal. Although the magnitudes of effects differ for other fissile compositions and other core shapes, the appropriate curves scale similarly to those of Fig. 6. We note that the reflectors that are much better than water at considerable thicknesses (beryllium, beryllium oxide, heavy water, graphite, uranium, and pure nickel) are uncommon materials except in certain reactors. Other than keeping in mind the unusual effects of these special reflectors, we assume that they need not be considered in normal operations.

Concrete, not represented in Fig. 6, is both common and a somewhat better reflector than water.¹⁸ It is unusual, however, to have concrete fitted closely about fissile material, and the customary concern is about the reflection effect of concrete when it is separated by a number of inches from the

object it influences. Lloyd and Clayton¹⁹ at Hanford measured the effect of a 6-in.-thick shell of concrete surrounding a 14-in.-diam sphere of plutonium solution but separated from it by a 4-in. gap. They observed that reflection by this concrete was about equivalent to that of a 1-in.-thick, close-fitting layer of water. The effect of a 12-in.-thick concrete wall against a 9-in.-diam cylinder of U(93) solution (330 g ^{235}U /liter, unreflected critical height 23 in.), even more like plant conditions, was investigated by Fox, Gilley, and Callihan²⁰ of ORNL. Their results indicate that reflection by the wall was less than that of a 0.2-in.-thick, close-fitting layer of water. When the wall is 6 in. away from the cylinder, its influence is reduced by another factor of 3 or 4, and so is negligible for practical purposes.

Steel, as used in forming dies or pressure vessels, and water are the principal reflector materials that are likely to be both close-fitting and thicker than a fraction of an inch. (Low-density thermal insulation is a poor reflector, so it is not in the same class.) Because of the pronounced effect of spacing, we conclude that structures of good reflector material such as concrete are hardly ever more effective than close-fitting water. Relative critical volumes of different fissile cores surrounded by various thicknesses of water are shown in Fig. A2.

It is apparent from Fig. 6 that water has unusual features as a reflector. At thicknesses up to an inch or so, it is among the more effective materials, but an increase of thickness beyond several inches adds little to its influence. (For shapes such as long cylinders or slabs, the "saturation" thickness is somewhat greater than that for the small spheres of Fig. 6.) This peculiar behavior of water is a consequence of neutron moderation and subsequent capture by hydrogen. After passage through several inches of water, the average neutron has so little energy that it is more likely to be captured by hydrogen than to find its way back to the fissile core.

We should make it clear that we use the term "water-reflected" here to imply that the fissile system of concern is closely surrounded by at least 6-in.-thick water. Thus the reflection effect is essentially the maximum attainable with water.

We stated earlier that fissile objects as well as inert materials may be viewed as neutron reflectors.

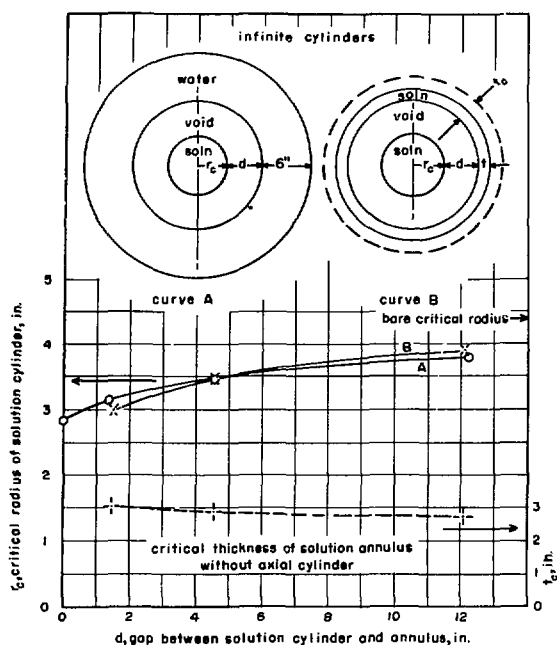


Fig. 7. Computed critical radius of solution cylinder vs distance to surrounding annulus of water or of solution. The cross section of the solution annulus is one-half that of the annulus which is critical without the axial cylinder (i.e., critical thickness $t_c \sim 2t$ at fixed $r_c + d$).

This point is illustrated by Fig. 7 which gives computed (DSN)²¹ critical radii of infinitely long cylinders of U(93) solution at 500 g of ^{235}U /liter. Curve A shows how the critical radius increases as a 6-in.-thick annulus of water is moved outward from the cylinder. Curve B is similar, except that a thinner surrounding annulus contains the same solution as the central cylinder. For each configuration, the cross section of the solution annulus is adjusted to be one-half that of the unreflected critical annulus of the same inside diameter (the dotted curve). For spacings greater than 2 in., the two curves are surprisingly similar. At a spacing of 7 in., the reflector saving of either annulus is just one-half that of close-fitting water.

Qualitatively, it is clear why a gap between a fissile core and surrounding material has such a pronounced effect. As spacing increases, the core simply becomes a poorer and poorer target for neutrons scattered back from the reflector. This picture also shows why the influence of a gap around a

spherical core is greater than that of the same gap about a corresponding long cylinder or thin slab of similar material. The sphere is the poorest target for returning neutrons, the cylinder is somewhat better, and the slab intercepts still more of the reflected neutrons.

MORE ABOUT SOLUTIONS AND MIXTURES

The homogeneous U(93) metal-water mixtures illustrated in Figs. 2 and 3 are, of course, not encountered in practice. The "experimental" values in these figures were derived by applying small corrections to results for uranyl-fluoride solutions, and these corrections become negligible for ^{235}U concentrations below 200 g/liter (or for volume fractions of uranium metal below 0.01). In other words, critical dimensions of hypothetical metal-water mixtures and of solutions coincide over the concentration range of most interest for aqueous processing. (See Figs. A3-A6 for U(93), Figs. A7-A10 for plutonium, and Figs. A11-A14 for ^{233}U .) However, as ^{235}U concentrations increase, particularly beyond the solution range, deviations of practical mixtures from metal-water become increasingly significant.

The fictitious mixture is used as a reference composition because its critical mass is minimum at a given water- ^{235}U ratio. This follows because the ^{235}U density is greater in the metal-water mixture than it is in a water mixture of any uranium compound at the same H/ ^{235}U atomic ratio.* Little experimental criticality information exists for the range of composition that includes aqueous slurries and filter cakes. Here, metal-water values may be applied conservatively for nuclear-safety evaluation.

Computed critical masses of several U(93)-water mixtures in the form of water-reflected spheres appear in Fig. 8. The abscissa represents ^{235}U density, and the curve at densities less than 0.8 kg of ^{235}U /liter applies to uranyl-fluoride solutions. At the right, the upper curve is for mixtures of UF_4 and water, the middle curve applies to UO_2 -water, and the lower curve gives metal-water reference values.

*The atomic ratios, H/ ^{235}U , H/Pu, and H/ ^{233}U are used as indexes of the degree of hydrogen moderation. Collectively, these ratios are frequently designated H/X, where X represents ^{235}U , Pu, or ^{233}U . Relations between H/X and density of X for certain solutions, metal-water mixtures, and some other mixtures are given in Table I and Fig. 12 of TID-7028.

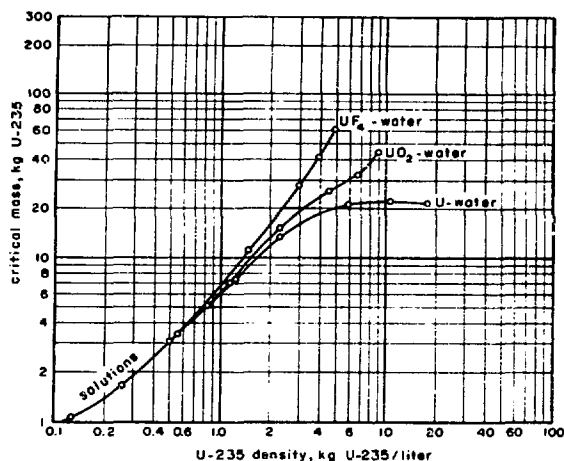


Fig. 8. Computed critical masses of water-reflected spheres of mixtures of water with U(93.5) metal, U(93.5)O₂, or U(93.5)F₄.

The principal purpose of this illustration is to show the penalty that can be paid if metal-water values are assumed for practical mixtures. But we should add that this penalty sometimes may be outweighed by the uncertainties of calculated results.

As the greatest differences in Fig. 8 are for dry (unmoderated) materials, it is instructive to examine a greater range of compounds under this condition. In Fig. 9, UC₂, UO₂F₂, and UF₆ (condensed) have been added to the three materials shown in Fig. 8. Critical masses apply to water-reflected spheres of the undiluted fissile compound at crystal density.²² The curve refers to unmoderated U(93) metal at various ²³⁵U densities. Departures of critical masses of compounds from the line show that the ²³⁵U density is not the whole story, and that the carbon, oxygen, and fluorine atoms have some influence on critical size. (Note that deviations from the curve are roughly proportional to the number of diluent atoms per uranium atom.) Although these atoms are too few to have a significant moderating effect, their nuclei do scatter neutrons so that the atoms near the outside of the core return toward the center some neutrons that would otherwise pass into the surrounding water. The effect is like that of an added internal reflector.*

*Part of the scattering effect of fluorine is compensated by capture of very high-energy neutrons such as occur in the unmoderated systems of Fig. 9.

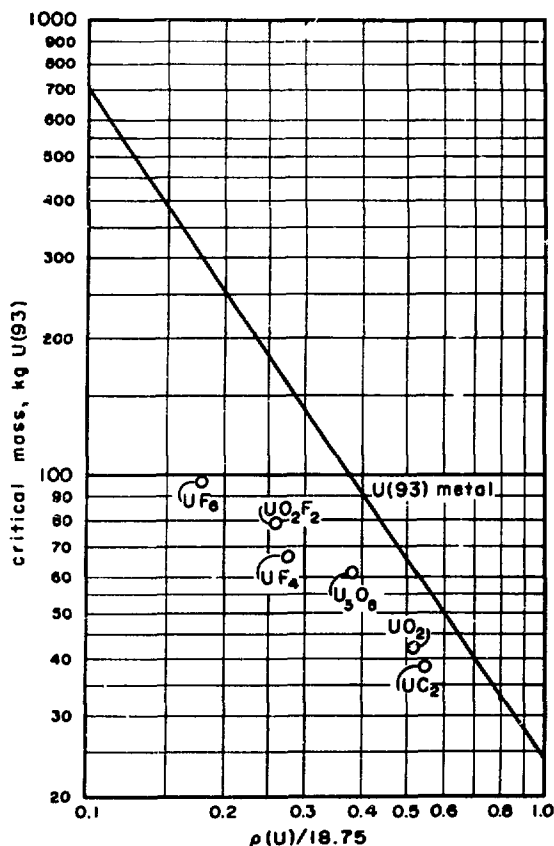


Fig. 9. Computed critical masses of water-reflected spheres of U(93) compounds. The line applies to U(93) metal at various fractions of normal density.

Measured effects of diluting U(93) and plutonium metal with other materials are given by Figs. A15 and A16, and by Ref. 23. Figure A16 is particularly interesting because it shows that carbon atoms must be present in large proportion to moderate neutrons significantly. The other effective moderating materials, deuterium, beryllium, and beryllium oxide, shown by this figure, are encountered almost exclusively as reactor components. (Graphite, of course, is also used for crucibles and molds in casting furnaces.)*

*Note that these moderating materials lead to small critical masses at very low ²³⁵U densities. (Deuterium, beryllium, carbon, and oxygen do not capture neutrons as readily as does hydrogen, so they do not poison very dilute ²³⁵U.) Although ²³⁵U critical masses may be small, the corresponding volumes are large.

The need to process reactor-fuel compositions such as U-graphite and U-BeO leads to concern about critical masses of the mixtures U-graphite-H₂O and U-BeO-H₂O. Excepting some data for U(93)-graphite-plastic compositions, there is no experimental information about these ternary mixtures. (Perhaps "fourfold mixtures" is the better term, because the uranium enrichment may be less than 93% ²³⁵U, which leads to ²³⁵U-²³⁸U-graphite-H₂O and ²³⁵U-²³⁸U-BeO-H₂O. The situation, of course, becomes even more complex when plutonium is built into the fuel.) In the particular case of U(93)-graphite-H₂O, there is a computational survey by Stratton^{5,24} of Los Alamos. His results for unreflected spheres are reproduced in Fig. 10. This family of curves illustrates the complex trade-off among effects of density change, scattering and moderation by graphite, and moderation and capture by hydrogen.

The power-reactor fuel that is presently most abundant is ²³⁵UO₂-²³⁸UO₂ (if fuel-cladding materials are ignored), which implies that criticality data for the ternary system ²³⁵UO₂-²³⁸UO₂-H₂O are of extremely practical interest. Appropriate to this combination are experimental critical dimensions of homogeneous mixtures of hydrogenous material and uranium at enrichments of less than 93% ²³⁵U, which appear in Figs. A1 and A17. (See Ref. 25 for a computational survey.) It is apparent from these data that ²³⁸U behaves like a neutron poison when it is present in much greater abundance than ²³⁵U. In fact, uranium containing less than 1.0% ²³⁵U cannot be made critical as a homogeneous mixture with water or other hydrogenous material.

HETEROGENEOUS URANIUM-WATER SYSTEMS

Of course, many nuclear-safety questions about the customary fuels for reactor lattices cannot be

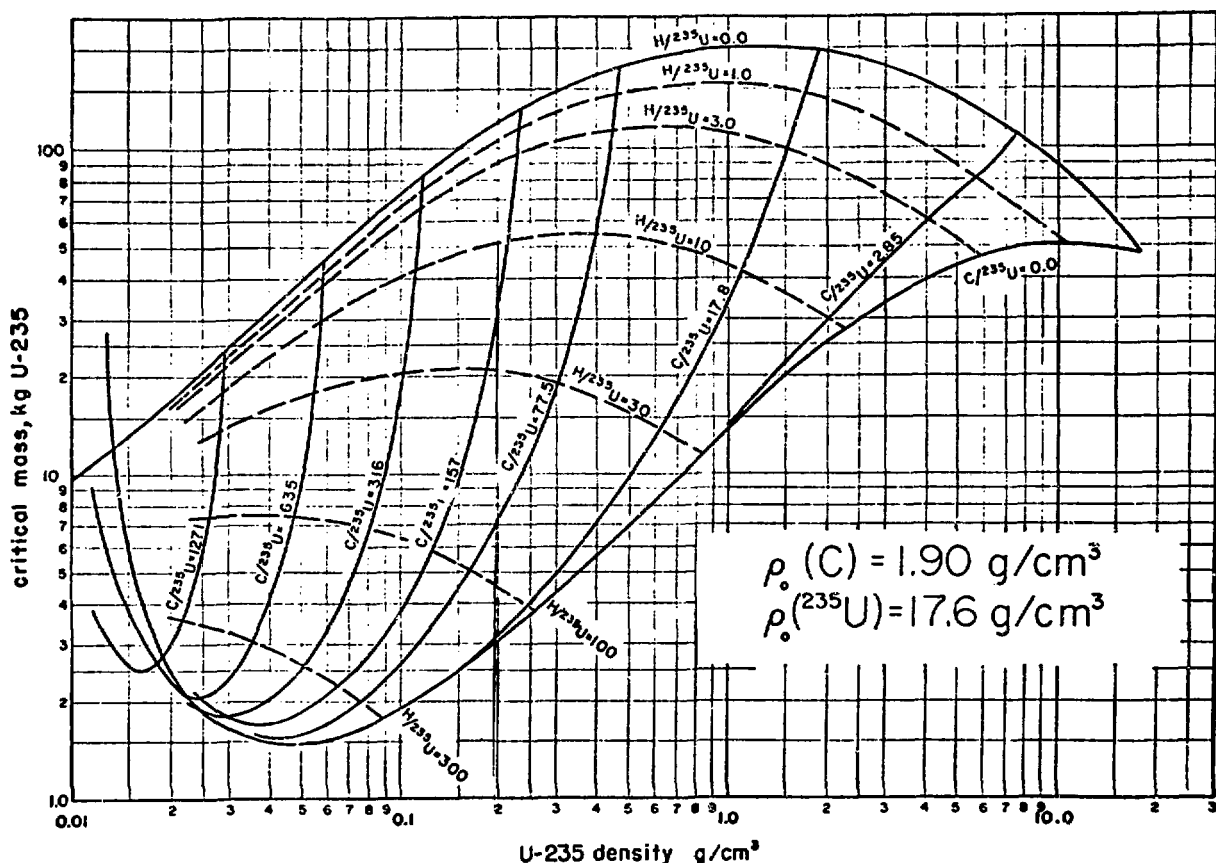


Fig. 10. Computed critical masses of unreflected spheres of U(93.5)-water-graphite mixtures.

answered by information about homogeneous mixtures. Certain fuel fabrication steps, storage, and transportation may involve regular lattices of fuel elements in water; dissolving the spent fuel for reprocessing may involve either regular lattices or random arrangements of chopped elements, but in either case there is a mixture of solids and solutions with changing composition. Considering the range of materials that may be represented in various fuel elements and processes (which can be non-aqueous), there are so many variables that criticality data cannot be mapped comprehensively. Specialized data required for a given fuel can be obtained most readily during development of the reactor for which the fuel is intended. If this is not done, there is the choice between applying very conservative nuclear-safety controls or obtaining specific criticality data. Depending upon circumstances, economy may dictate either alternative.

With the above choice in mind, it behooves us to have some idea about how to establish "very conservative" controls. Most of the fissile material in present power-reactor fuels in the United States is uranium with ^{235}U enrichments of 2 to 5%. (We refer to fuels for boiling-water and pressurized-water reactors.) For this class of fuel, a basis for conservative criticality control is the considerable experimental information about lattices of slightly enriched uranium rods immersed in water.

One set of conclusions from measurements with latticed uranium in water is summarized in Fig. A18. In this figure, minimum critical masses of lattices and of homogeneous mixtures (both water-reflected) are compared over a range of ^{235}U enrichments. Each point on the lattice curve implies optimum rod diameter, optimum spacing between rods, a near-spherical lattice shape, and complete water reflection. The feature of this curve is that it falls below the curve for homogeneous spheres at ^{235}U enrichments of less than ~ 5%. Above about 5% ^{235}U , minimum critical masses occur for the homogeneous systems (which may be viewed as lattices in which rod diameter is zero).

Our qualitative picture of neutron behavior helps us to understand the reduced critical masses of lattices at low enrichments. As mentioned before, neutron capture by ^{238}U (its "poison" effect) drives the homogeneous curve to infinity at 1.0% ^{235}U . It

happens that neutrons of intermediate energy (partially moderated) are captured more readily by ^{238}U than are either high-energy fission neutrons (unmoderated) or neutrons of lowest energy (fully moderated or "thermal"). In a homogeneous system, neutrons are exposed continually to capture by ^{238}U while being moderated. In a lattice, however, the typical neutron from a fission in one rod travels through water that is free of ^{238}U before reaching another rod (or being returned). At "optimum" lattice geometry, this average path through water is such that some neutrons, which otherwise would be captured by ^{238}U , are moderated sufficiently to escape this fate. Thus, fissions are produced more efficiently than if the system were homogeneous, so that the critical mass is smaller.

Now, various stages of conservatism in the criticality evaluation of fuel-rod lattices in water become apparent.

- The extreme of conservatism would be to apply minimum criticality data for highly enriched uranium (homogeneous systems).
- The minimum critical mass of all possible lattices at the appropriate ^{235}U enrichment may be used. Alternatively, the minimum critical dimensions may be chosen from Figs. A19-A21.
- The guiding criticality data may be derived from uranium-metal lattices similar to the lattices of actual fuel. For example, they may be obtained from the sources of the information summarized in Fig. A18.
- Data may be specific to the fuel-element lattice of interest.

A generalization falling between the last two alternatives is suggested by existing experimental information for specific power-reactor fuel elements. Figure 11, for example, applies to water lattices of elements that were studied for use in pressurized-water reactors.^{26,27} The lower curve of minimum critical volume vs ^{235}U enrichment is for unclad uranium-metal rods and should be somewhat conservative for zirconium-clad UO_2 rods. The two points above are for stainless-steel-clad UO_2 . Because of the relatively small ranges of fuel-element dimensions that are suitable for water-moderated power reactors, the data of Fig. 11 will not be in great error if applied to any rod-type $\text{U}(2 \text{ to } 5)\text{O}_2$ element with the appropriate cladding material. There are

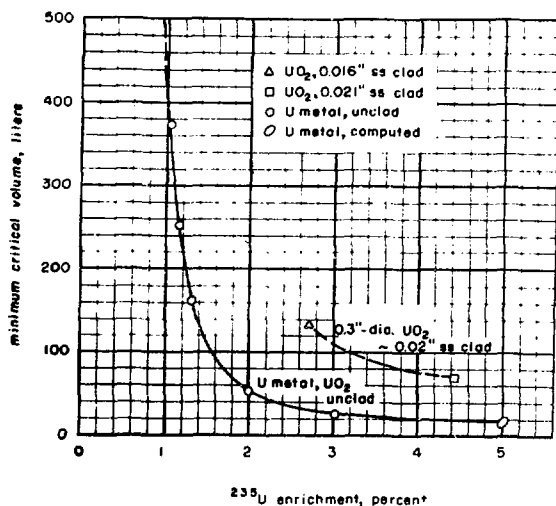


Fig. 11. Minimum critical volumes of water lattices of low-enrichment uranium or UO_2 rods, as water-reflected spheres.

no experimental data about corresponding lattices in solutions of slightly enriched uranium, such as might be encountered in pickling baths or fuel dissolvers.

Before leaving water lattices, let us consider the data for U(94)-metal lattices shown in Fig. A22. This figure indicates a large reduction of attainable critical mass as pieces of the highly enriched metal become smaller. Another item may be mentioned: The only experimental evidence about combinations of fissile solids and fissile solutions applies to U(93), and is represented by Figs. 25 and 26 of TID-7028 and by references in the associated text.

MORE ABOUT POISONS

As we have mentioned, hydrogen and ^{238}U capture neutrons readily enough to behave like mild neutron poisons (^{240}Pu is similar). Of the elements in common uranium and plutonium compounds, carbon, oxygen, and fluorine have negligible poisoning effect, nitrogen is a mild poison,* and chlorine is a moderate poison. Among structural materials, aluminum has a small neutron-capture effect, copper and the components of steels are mild poisons, and glass is influenced by its boron content.

* This is why critical dimensions of uranyl-fluoride solutions are slightly smaller than those of uranyl-nitrate solutions.

One method of controlling solution criticality, mentioned in the discussion of accident experience, is to add strong poisons to the fissile material. The neutron poisons most suitable for this purpose are boron, cadmium, and the rare earths, samarium, europium, and gadolinium. Cadmium's strong capture effect is limited to neutrons of very low energy (highly moderated), so this material is most useful for dilute aqueous solutions. As the other strong poisons remain effective for neutrons of intermediate energy, they are more generally applicable. Of course, boron is preferred over the rare earths because of its much lower cost. There is no very effective poison for the unmoderated neutrons that are typical of undiluted fissile metal and most dry compounds.

Of the strong poisons mentioned, the experimental data of TID-7028 (pp. 46-48) apply exclusively to boron. Three types of system are represented: homogeneous mixtures such as soluble boron in uranium solution, boron-containing solids distributed throughout uranium solution, and boron solution in lattices of fuel rods. In most cases the quantity of boron required to prevent criticality in any finite system is established. This is accomplished if the reproduction factor of the infinite system, k_∞ , is unity.

The rather scant experimental information about homogeneous mixtures may be supplemented by the (DTK) computed curves of Fig. 12.²⁸ Atomic ratios B/X ($X \equiv ^{235}\text{U}$, ^{239}Pu , or ^{233}U) for which $k_\infty = 1$ are shown

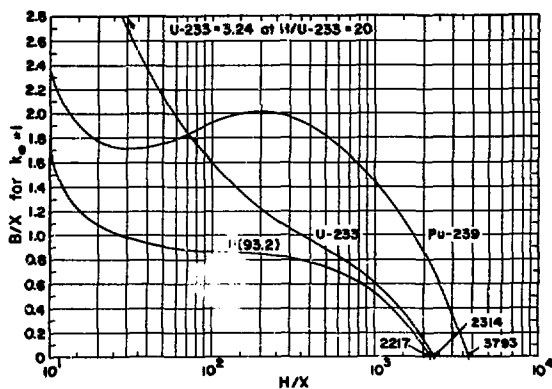


Fig. 12. Computed atomic ratios of boron to fissile isotope at which $k_\infty = 1$, for mixtures of water with U(93.2), ^{235}U , or ^{239}Pu (in the atomic ratios B/X and H/X , where $X \equiv ^{235}\text{U}$, ^{239}Pu , or ^{233}U).

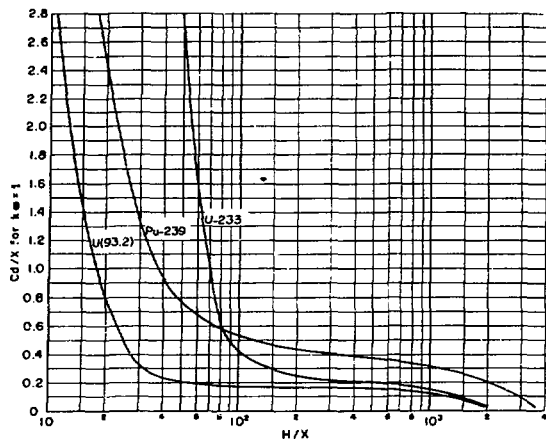


Fig. 13. Computed atomic ratios of cadmium to fissile isotope at which $k_{\infty} = 1$, for mixtures of water with U(93.2), ^{233}U , or ^{239}Pu (in the atomic ratios Cd/X and H/X , where $X \equiv ^{235}\text{U}$, ^{233}U , or ^{239}Pu).

for solutions of U(93), ^{239}Pu , and ^{233}U . Qualitatively, a greater proportion of boron is required for ^{239}Pu and ^{233}U solutions than for ^{235}U , to overcome the more efficient fissioning of these materials. Figure 13 shows results of similar calculations for cadmium-poisoned solutions. The effect of cadmium decreases rapidly as the solutions become so concentrated that very low-energy neutrons disappear.

A convenient means of poisoning a solution is by packing the container with raschig rings of borosilicate glass. A tabulation of experimental conditions for which $k_{\infty} = 1$ with glass in ^{235}U solutions appears on p. 47 of TID-7028. To illustrate an influence of the heterogeneous distribution of boron, that table may be extended as in Table IV. The listed overall $B/^{235}\text{U}$ ratios for $k_{\infty} = 1$ are expected to be greater than corresponding values from Fig. 12,

TABLE IV

CONDITIONS AT WHICH $k_{\infty} = 1$ WITH ≤ 1.5 -in.-o.d. GLASS RASCHIG RINGS IN U(93) SOLUTION

| Volume Fraction of Glass | Boron Content of Glass (wt %) | ^{235}U Content of Solution | | Atomic Ratio $B/^{235}\text{U}$ |
|--------------------------|-------------------------------|---|--------------------|---------------------------------|
| | | $\text{g } ^{235}\text{U}/\text{Liter}$ | $H/^{235}\text{U}$ | |
| 0.24 | 3.3 | 385 | 64 | 1.9 |
| 0.22 | 4.0 | 385 | 64 | 2.0 |
| 0.24 | 0.5 | 67 | 390 | 1.6 |

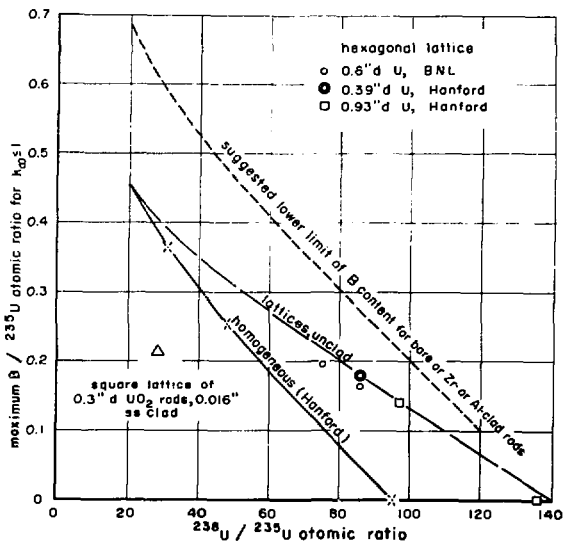


Fig. 14. Atomic ratios of boron to ^{235}U at which $k_{\infty} = 1$ for water-moderated uranium at enrichments of less than 5% ^{235}U .

because boron when lumped, as in the glass rings, is less effective than when distributed uniformly. Boron captures low-energy neutrons so well that its effect "saturates" as thickness is increased, which leads to greater capture per atom in small thicknesses (or a homogeneous distribution) than in greater thicknesses.

A further remark about fixed poisons such as glass rings or boron-stainless grids is that their effectiveness diminishes as the cell size or spacing increases beyond about 1 in. This influence for parallel boron-steel plates in U(93) solution is illustrated by the last entry in Table 5 of TID-7028.

For uranium of low ^{235}U enrichment, it is expected that k_{∞} will be unity at smaller $B/^{235}\text{U}$ atomic ratios than indicated by Fig. 12. This difference is shown by experimental data for U(5) that are summarized in Fig. 14. The lower curve applies to homogeneous boron-poisoned mixtures, and the upper solid curve is for lattices of rods with boron dissolved in the intervening water.

IV. CRITICALITY INFORMATION—LOW-DENSITY UNITS AND ARRAYS

Homogeneous critical systems at low density are of interest primarily as limiting cases of arrays of units separated by air. For example, in a dry-storage

arrangement of fuel bundles for water-moderated reactors, each fuel element is usually so small that the entire array behaves like an assembly containing the same materials spread uniformly throughout the same overall volume. If the fuel were to be lumped into progressively larger elements, the behavior would be less like that of the homogeneous assembly with the same average material densities. Nevertheless, we can still expect certain similarities between arrays of large units and homogeneous systems at low densities.

If such similarities can be found, the well-known influence of density on the critical size of an individual core may suggest useful empirical forms for correlating critical data for air-spaced arrays. The required application, of course, is to the criticality evaluation of ever-present air-spaced storage arrays and stacks of packages of fissile material.

HOMOGENEOUS LOW-DENSITY SYSTEMS

An intriguing aspect of our qualitative picture of neutron behavior is that it leads to one exact quantitative relationship that applies to any critical assembly in which the density is changed uniformly. If all dimensions of an assembly are scaled inversely as the density, any neutron path from one region to another scales in the same way, and the number and kinds of nuclei along this path remain unchanged, so there is no change in neutron processes. In other words, the relative numbers of neutrons producing fission, being captured, being scattered, and leaking from the system are not changed, so the assembly remains critical. Thus, critical dimensions are inversely proportional to the density, provided the density changes are uniform. For a reflected system in which the densities of core and reflector are changed by the same ratio, this implies that critical dimensions of both core and reflector scale inversely as the density. (If only the reflector density or only the core density changes, the above relationship no longer applies to the core dimensions.)

Where all densities are changed by the ratio ρ/ρ_0 , it follows that any critical dimension ℓ_c is given by

$$\frac{\ell_c}{\ell_{co}} = \left(\frac{\rho}{\rho_0}\right)^{-1} \quad (5)$$

where ℓ_{co} applies to the initial density ρ_0 . As core and reflector densities are seldom changed in the same proportion, this expression is most commonly applied to unreflected fissile material.

For a bare spherical core of initial critical radius, r_{so} ,

$$\frac{r_s}{r_{so}} = \left(\frac{\rho}{\rho_0}\right)^{-1}, \quad (6a)$$

so the relation for critical volume is

$$\frac{V_s}{V_{so}} = \left(\frac{\rho}{\rho_0}\right)^{-3} \quad (6b)$$

and the critical mass ratio is

$$\frac{m_s}{m_{so}} = \left(\frac{\rho}{\rho_0}\right) \frac{V_s}{V_{so}} = \left(\frac{\rho}{\rho_0}\right)^{-2}. \quad (6c)$$

For a bare, infinitely long cylinder of initial critical radius r_{co} , we have

$$\frac{r_c}{r_{co}} = \left(\frac{\rho}{\rho_0}\right)^{-1}. \quad (7a)$$

The expression for critical volume per unit length is

$$\frac{V_c}{V_{co}} = \left(\frac{\rho}{\rho_0}\right)^{-2}, \quad (7b)$$

and critical mass per unit length is

$$\frac{m_c}{m_{co}} = \left(\frac{\rho}{\rho_0}\right)^{-1}. \quad (7c)$$

If the shape of interest is an unreflected infinite slab of initial critical thickness, t_0 , again

$$\frac{t}{t_0} = \left(\frac{\rho}{\rho_0}\right)^{-1}. \quad (8a)$$

The critical volume per unit surface area varies in the same manner; in other words,

$$\frac{V}{V_0} = \left(\frac{\rho}{\rho_0}\right)^{-1}. \quad (8b)$$

Finally, the critical mass per unit area remains unchanged, or

$$\frac{m}{m_0} = \text{constant}. \quad (8c)$$

Of more practical interest than the above expressions, is the variation of critical dimensions of a reflected core when the reflector density remains constant. The small amount of available

experimental information about density changes in near-homogeneous cores is presented in TID-7028, p. 4 and Fig. 6. Results are consistent with a relationship like Eq. (6c) for critical masses of spheres (or cubes, or near-equilateral cylinders), so

$$\frac{m_s}{m_{s0}} = \left(\frac{\rho}{\rho_0} \right)^{-s}, \quad (9)$$

where s is approximately constant over the range $0.5 \leq \rho/\rho_0 \leq 1$. (The exponent s cannot exceed the value 2 that applies to a bare sphere.) Actually, s for a core with constant-density reflector must increase with decreasing density. As core density approaches zero, core size approaches infinity and s must approach 2, because the distinction between infinite bare and reflected cores is meaningless.

Figure 15 gives (DTK) calculated values of the core-density exponent s for a variety of water-reflected spheres, which apply to the density range between normal and 0.8 normal. The value $s = 1.5$ is typical of small cores, but s generally increases with core size and approaches 2 as the core volume becomes very large.

Computations (DTK)²² that extend to very small values of ρ/ρ_0 , Figs. 16 and 17, show the increase of the core-density exponent of a water-reflected sphere with decreasing density. The exponent becomes essentially 2 when $\rho/\rho_0 \leq 10^{-3}$. At these very low densities, all curves are parallel and there is a constant ratio of critical masses of bare and

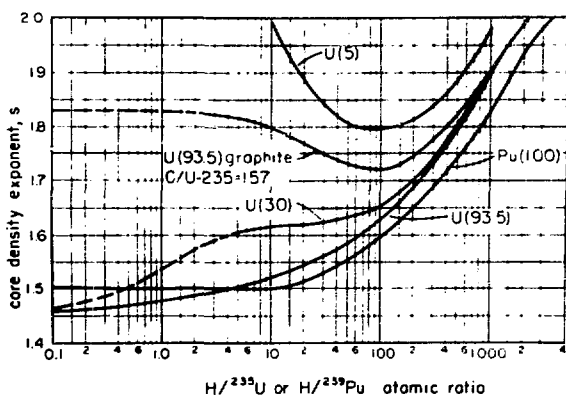


Fig. 15. Computed initial core-density exponents for water-reflected spheres of mixtures of water with ^{239}Pu , $\text{U}(93.5)$, $\text{U}(30)$, $\text{U}(5)$, or $\text{U}(93.5)$ -graphite at $\text{C}/^{235}\text{U} = 157$. Critical sizes are infinite where $s = 2$.

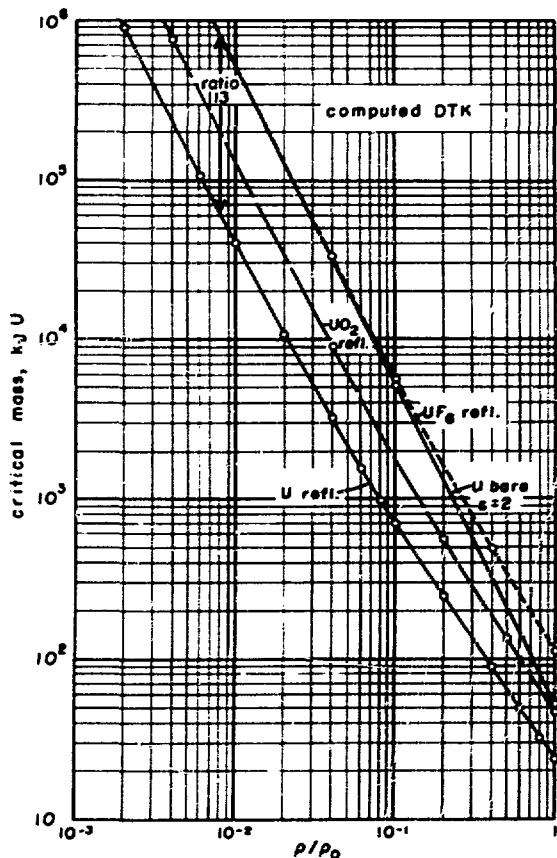


Fig. 16. Computed critical masses of spheres with core densities reduced by the fraction ρ/ρ_0 ; water-reflected $\text{U}(93)$ metal, $\text{U}(93)\text{O}_2$, or $\text{U}(93)\text{F}_6$, and unreflected $\text{U}(93)$ metal.

corresponding reflected spheres. Values of this maximum ratio, R , for the various core compositions of Figs. 16 and 17, are shown in Table V.²⁹

TABLE V
LIMITING RATIOS OF CRITICAL MASSES OF BARE AND WATER-REFLECTED SPHERES AT LOW DENSITY

| Core Composition | $m_s(\text{bare})/m_s(\text{refl})$ |
|--|-------------------------------------|
| $\text{U}(93)$ metal | 13 |
| $\text{U}(93)\text{O}_2$ | 8.0 |
| $\text{U}(93)\text{F}_6$ | 6.0 |
| $\text{U}(93)\text{C}_{80}$ | 2.7 |
| $\text{U}(93)$ solution, $\text{H}/^{235}\text{U} = 60$ | 5.4 |
| $\text{U}(93)$ solution, $\text{H}/^{235}\text{U} = 400$ | 2.7 |
| Pu metal | 19 |

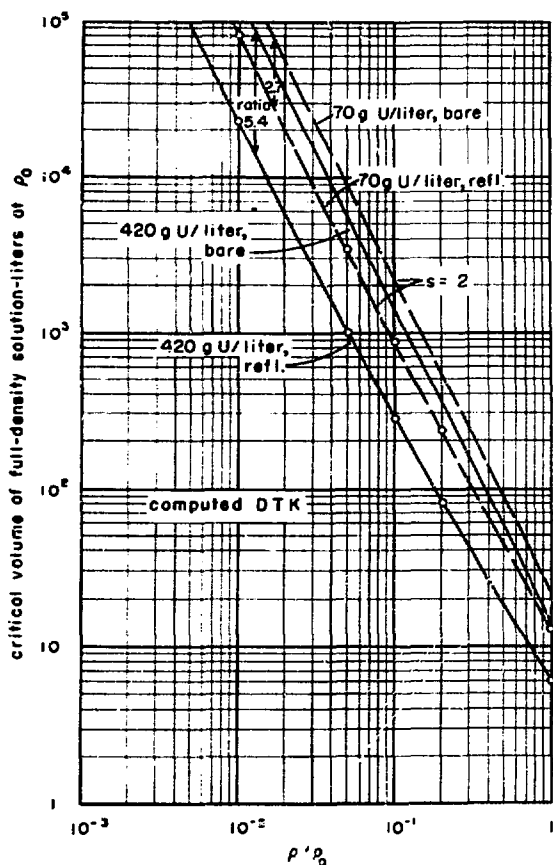


Fig. 17. Computed relative critical masses of spheres with core densities reduced by the fraction ρ/ρ_0 ; water-reflected and unreflected U(93) solutions at 70 or 420 g U/liter ($M/^{235}\text{U} = 400$ or 60).

As indicated before, the core-density exponents for homogeneous systems may be applied to certain materials that are actually heterogeneous, such as oxide pellets, machine turnings, and dry lattices of fuel rods for water-moderated reactors. Heterogeneity will be unimportant if each piece is so small that a neutron originating in it has little chance of producing another fission before leaving the piece.* For many materials and shapes, this requirement will be satisfied if one dimension of the piece does not exceed approximately 1/2 in.

* An equivalent statement is that the "neutron multiplication" of the piece should be little greater than unity.

NEAR-EQUILATERAL AIR-SPACED ARRAYS

We can expect the critical mass of a low-density system to decrease as the fissile material is lumped into larger units while the same overall density is maintained. In other words, the critical mass of a three-dimensional air-spaced array is smaller than that of a similar homogeneous system of the same material at the same average density. As more material is lumped into units, there is increased chance that a fission within one unit will lead directly to other fissions within the same unit, and the fission-chain efficiency of the entire array also increases. In the limit, the most efficient "array" (that of smallest critical mass) consists of one critical unit at full density.

To illustrate how the critical content of arrays depends upon unit size and the spacing between units (overall density), we turn to precise experimental information about arrays which appears in Part II of TID-7028.* Each of the critical air-spaced arrays to be considered has the same number of U(93) metal or solution cylinders along each of the three principal axes. In other words, each array contains 8, 27, 64, ... units. Cylinders are compact in that their heights do not differ greatly from their diameters, and the surface-to-surface spacing between cylinders is uniform. Reflected arrays are surrounded by 1- to 6-in.-thick paraffin spaced from the outer units by one-half the surface-to-surface spacing within the lattice. A metal array, as set up at ORNL, is shown in Fig. 18.

Data for critical arrays of U(93) metal cylinders are shown in Fig. 19, and Fig. 20 gives similar data for cylinders of solution in 1/4-in.-thick Plexiglas containers. The abscissa $\bar{\rho}/\rho_0$ is the fraction of the lattice volume occupied by U(93) metal or solution (the lattice volume per unit). The reflected arrays so represented are surrounded by 6-in.-thick paraffin.

For criticality safety guidance, the unreflected arrays are of only academic interest because practical storage arrangements always involve some reflection, often by concrete walls. Although concrete is

* All of the suitable criticality data for arrays in TID-7028 originated at ORNL. Other information about subcritical arrays can be valuable for evaluating the safety of similar systems, but it is not appropriate for developing a general model.

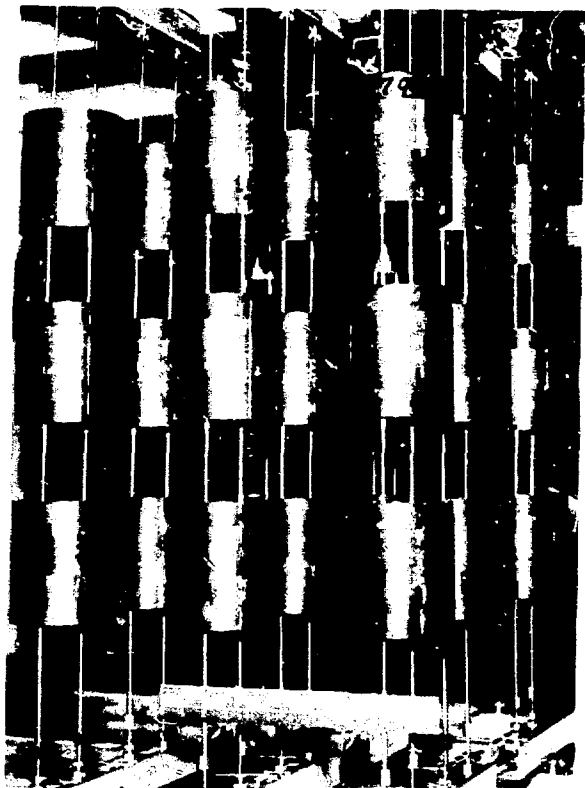


Fig. 18. Unreflected air-spaced array of 26.1-kg cylinders of U(93) metal (described on p. 133 of TID-7028).

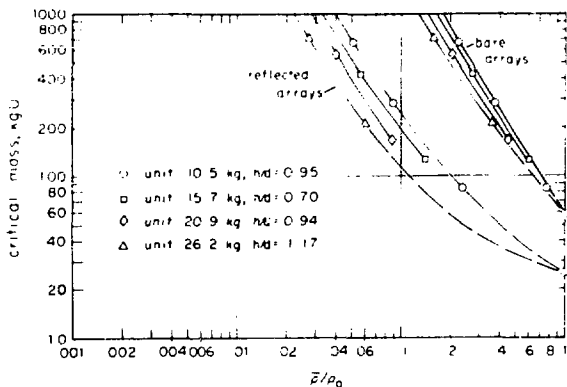


Fig. 19. Critical masses of air-spaced arrays of 10.5-, 15.7-, 20.9-, or 26.2-kg cylinders of U(93) metal. There is the same number of units along each of the three principal axes; surface-to-surface spacing is uniform.

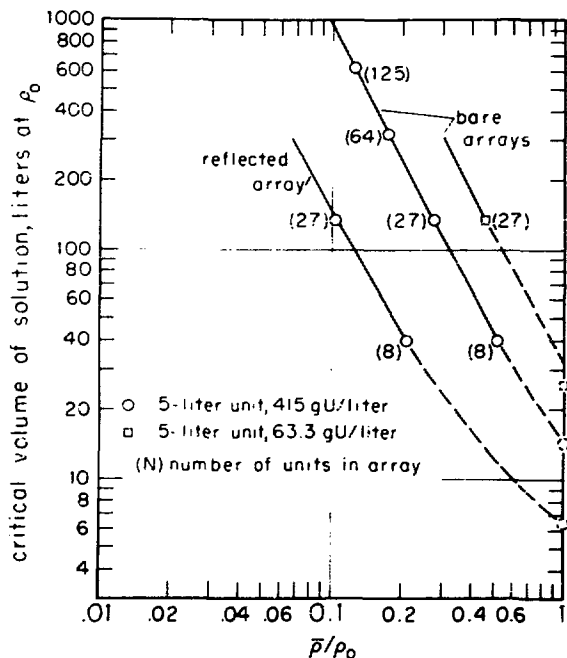


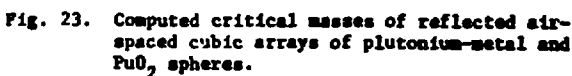
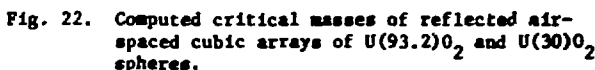
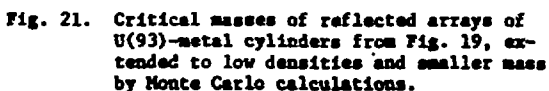
Fig. 20. Relative critical masses of air-spaced arrays of near-equilateral U(93)O₂(NO₃)₂ solution cylinders. There is the same number of units along each of the three principal axes; surface-to-surface spacing is uniform.

a better reflecting material than paraffin, the close-fitting, 6-in.-thick paraffin box is more effective than the usual concrete vault with passageway and some walls at a distance from stored units. Thus the data on reflection serve as a reference for evaluating the safety of storage arrays of similar units. (Effects of containers and other materials, such as water, which may enter the lattice are discussed later.)

Similarities among Figs. 19 and 20 and the curves of Figs. 16 and 17 for homogeneous systems had suggested the so-called "density-analog" technique for estimating critical sizes of arrays.^{29,30} Briefly, a relation like Eq. (9) with a conservatively chosen value of the exponent s was used to extrapolate from a single bare critical unit to a bare array at the desired value of $\bar{\rho}/\rho_0$. Then a reflection ratio from Table V was applied to give the size of the corresponding reflected array. (It is apparent that the curves for reflected arrays cannot be approximated directly by Eq. (9) with constant s .) Although this relatively crude approximation was useful

An extensive computational survey of arrays of many types of units has been conducted by J. T. Thomas of the Oak Ridge Critical Experiments Facility.^{31,32} For this purpose, he has used Monte Carlo data validated by experimental information (including that of Figs. 19 and 20), and a realistic extrapolation formula validated by both experimental and Monte Carlo data. The resulting extension of experimental data for highly reflected arrays of U(93) units is shown in Fig. 21. Similar results for spherical units of U(93)O₂, U(30)O₂, α -phase Pu,³³ and PuO₂ appear in Figs. 22 and 23. Other available families of idealized arrays include spherical units of U(>30)O₂, ²³³U metal, ²³³UO₂, and oxides with some moisture content. Further curves apply to arrays of units in packages suitable for transportation or storage.

Further, Thomas has made a comprehensive study of effects of departures from the idealized U(93) arrays. ³⁴⁻³⁶ He has examined influences upon



critical-array size of unit-shape changes and density changes, of intermingling different units, and of interposing materials such as steel, plastic, or water within the lattice. This information is a good guide for translating from the idealized arrays to practical storage configurations.

For example, let us consider the vault storage of as many as two hundred 10.5-kg hemispherical shells of U(93), each in a closed, 1/8-in.-thick-wall steel can on sturdy steel shelving. A safe spacing between such units might be judged as follows by reference to the curve for 10.5-kg cylinders in Fig. 21. This curve says that 2100 kg of the cylinders (the 200 units) would be critical at the average lattice density $\bar{\rho}/\rho_0 = 0.025$. The same number of shells at this lattice density would be subcritical because a neutron within a shell is more likely to escape the shell without producing more neutrons by fission than is a neutron in the near-equilateral cylinder. Let us say (as is probable) that Thomas' guidance indicates that part, but not all, of this effect is compensated by the presence of steel cans and shelving. (Further, Thomas shows that the steel cans prevent critical-mass reduction by water flooding.) Although it appears that the storage arrangement would be subcritical at the 0.025 lattice density, let us reduce critical mass by the seemingly liberal safety factor of 10. This leads to the 21,000-kg point on the curve of Fig. 21, which corresponds to $\bar{\rho}/\rho_0 = 0.0074$, the fraction of the lattice occupied by U(93). The 10.5 kg of metal has a volume of 34 in.³, so the space to be allowed per shell is $34/0.0074 = 4600$ in.³ or a 16.6-in. cube. Note that the safety factor of 10 was equivalent to increasing the center-to-center spacing of units from 11.1 to 16.6 in. If one wished to round off the spacing to 16 in., a usually adequate safety factor of greater than 8 would be retained. It is apparent that other ≤ 10.5 -kg metal units, such as cans of dry metal turnings or clusters of small pieces, could safely replace shells in the storage configuration.

Of course, the convenient matching of masses in this illustration is not essential. If, for example, we had considered 14-kg shells, either the reference curve for 15.7-kg cylinders could have been used directly, or another could have been obtained by interpolation between the curves for 10.5- and 15.7-kg

units. When appropriate families of reference data are not available, it may be desirable to use Monte Carlo techniques, such as KENO, explicitly for the realistic analysis of proposed storage arrays.

The solution arrays of Fig. 20 do not apply directly to most large storage arrangements because the near-equilateral containers are less practical than long cylinders of larger volume. Specific guidance, however, is available in the form of many experimental critical patterns of U(93) solution in tubular containers described in Part II of TID-7028. Similar results for $U(4.9)O_2F_2$ solution have been reported subsequently.³⁷

One extensive series of experiments established critical arrays of $U(93)O_2(NO_3)_2$ solution in a common type of polyethylene storage container. Each -0.35-in.-wall, 4.7-in.-diam, 44-in.-high vessel contained 12-3/4 liters of solution. From the many essentially unreflected arrays of these cylinders, three with equilateral outline can be represented on a plot like Fig. 20. One is a two-high arrangement like that shown in Fig. 24. The other two are one-high with square and hexagonal patterns. Figure 25

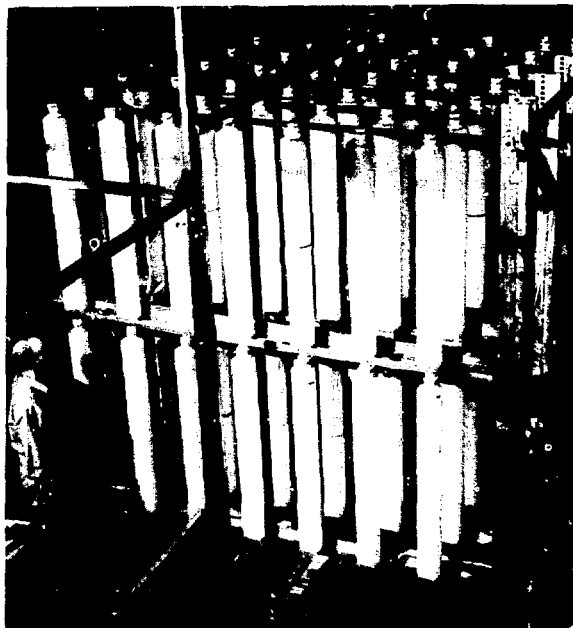


Fig. 24. Double-tier array of 13-liter polyethylene cylinders containing $U(92.6)O_2(NO_3)_2$ solution (described on pp. 80 and 111 of TID-7028.)

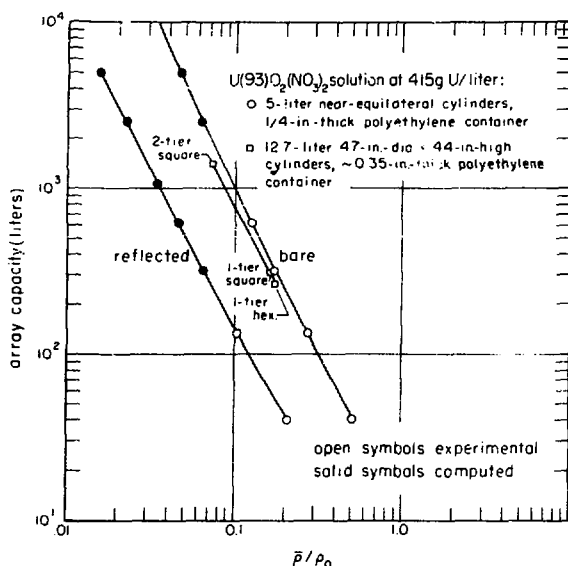


Fig. 25. Relative critical masses of near-cubic arrays of solution cylinders; extension of data shown in Fig. 20, and described on p. 111 of TID-7028.

is a computed extension of Fig. 20 with these three arrays of storage cylinders also represented. Because of the similarity of the "unreflected" curves, we may conclude that the curve for reflected arrays of 5-liter units also applies to storage cylinders if it is scaled down slightly. Note that the forest of long cylinders to which this generalization applies is not a common arrangement for solution storage. The safety of more usual layouts, such as a row of cylinders against a wall, may be judged

better by direct comparison with data from TID-7028, from the surface-density correlations we describe later, or from results of explicit Monte Carlo calculations.

As spacings between units do not appear directly on Figs. 21-23 and 25, specific illustrations of how overall critical mass depends upon spacing may be helpful. The curves of these figures lead to the numbers in Table VI.

Table VI shows first, that the critical spacings are not very large, and, second, that relatively small changes in spacing cause large changes in the critical content of an array. More specifically, an increase of about 15% in spacing doubles capacities of the first five listed arrays, and 50-60% increases capacities tenfold. The corresponding spacing increases for the long solution cylinders are ~ 25% and ~ 100%. The significance of this sensitivity to spacing is that a few extra inches between containers in a storage area can add a worthwhile safety margin, and, conversely, that dropping only slightly below a minimum acceptable spacing can be dangerous. It follows that items in storage arrays should be located by some positive means, not by eye, unless spacings are extremely conservative.

SURFACE-DENSITY MODEL OF ARRAYS

A surface-density "rule of thumb" is convenient for distinguishing clearly subcritical arrays of fissionable material from others that may require closer examination. This rule is easily applicable to many process arrangements in which each unit is substantially subcritical.

TABLE VI
CRITICAL SPACINGS FOR SELECTED TOTAL ARRAY CAPACITIES
OF WATER-REFLECTED NEAR-CUBIC ARRAYS

| Type of Unit | Av Center-Center (or Axis-Axis) Distance for Array Capacities | | |
|--|--|----------------------|----------------------|
| | in kg U(93) or Liters | 1000 kg or Liters | 5000 kg or Liters |
| 7.3-kg U(93), Fig. 21 | 6.8 | 7.8 | 10.3 |
| 10.5-kg U(93), Fig. 21 | 8.2 | 9.5 | 12.9 |
| 15.7-kg U(93), Fig. 21 | 10.0 | 11.6 | 15.7 |
| 20.9-kg U(93), Fig. 21 | 11.6 | 13.4 | 18.0 |
| 5-liter U(93) solution, Fig. 25 | 18.1 | 20.4 | 27.0 |
| Long, 4.7-in.-diam U(93) solution, Fig. 25 | -18 axial | -22 axial | -35 axial |

TABLE VII
REFERENCE SURFACE DENSITIES FOR SELECTED FISSIONABLE MATERIALS

| Composition | Maximum Unit Size | Reference Surface Density |
|---|-----------------------------|--|
| U(93) solution | 5.0-in.-o.d. cylinder | 1.6 m _l /cm ² (1.5 l/ft ²) |
| | 5.5-in.-o.d. cylinder | 1.4 m _l /cm ² (1.2 l/ft ²) |
| | 6.0-in.-o.d. cylinder | 1.2 m _l /cm ² (1.1 l/ft ²) |
| U(93) solution, ≤ 50 g U/t | 6.5-in.-o.d. cylinder | 3.6 m _l /cm ² (3.4 l/ft ²) |
| U(5) solution | 8.0-in.-o.d. cylinder | 9.6 m _l /cm ² (9.0 l/ft ²) |
| Stable Pu(NO ₃) ₄ solution | 4.7-in.-o.d. cylinder | 1.4 m _l /cm ² (1.3 l/ft ²) |
| ²³³ U solution | 4.5-in.-o.d. cylinder | 1.3 m _l /cm ² (1.2 l/ft ²) |
| U(93) metal | 15 kg U ^a | 13 g/cm ² (12 kg/ft ²) |
| U(93)O ₂ | 27 kg U ^a | 12.5 g/cm ² (11.5 kg/ft ²) |
| U(93)F ₆ | 50 kg U ^a | 11.5 g/cm ² (10.5 kg/ft ²) |
| U(93)F ₆ + 0.1HF | 50 kg U ^a | 7.5 g/cm ² (7 kg/ft ²) |
| α -phase Pu | 3 kg Pu ^a | 5.5 g/cm ² (5.2 kg/ft ²) |
| δ -phase Pu, or ²³³ U metal | 4.5 kg Pu or U ^a | 5.5 g/cm ² (5.2 kg/ft ²) |

^aReduced 5% for container effects.

The simplest situation is a uniform array of fissionable units. Then if the surface density of fissionable material, as projected onto the largest bounding plane of the array (usually floor or wall), does not exceed a reference value for that material, no more detailed evaluation is necessary. Reference surface-density values and corresponding unit-size limits for selected materials are listed in Table VII. As an illustration, consider 12-kg units of highly enriched uranium metal, arranged on the floor of a storage area in a square array with 15-in. center-to-center spacing. Table VII quickly shows that this configuration is acceptably subcritical, because the 12-kg unit is less than the 15-kg limit for U(93) metal, and the surface density, $12/(12/15)^2 = 7.7 \text{ kg/ft}^2$, is about two-thirds the reference value. Other simple situations to which the table applies would include a square array of vertical solution cylinders with surface density as projected onto the floor, or a line of storage cylinders along a wall with surface density as projected onto that wall.

The rule can be extended directly to nonuniform plant layouts in which surface densities for the various materials are separable (e.g. where, in a basically horizontal layout, different materials are not stacked in the same column). In such a case, the most intensive concentration of each material should be examined in terms of the values in Table VII. (As we shall see, some "nonseparable" cases can also be handled.) Although practical arrangements

seldom exceed the reference values, it should be emphasized that a larger surface density does not necessarily indicate an unsafe condition. Instead, it would call for a more detailed evaluation. It is apparent that the listed values of both surface density and unit size, which apply to infinite arrays, may be relaxed for finite systems. The extent of such relaxation, however, is a complication that we will not pursue.

To gather some insight, we should consider the way in which the surface-density rule is based on the properties of critical planar arrays of identical units, e.g. a square horizontal pattern, one unit high. As the array size is increased by adding units horizontally, the critical spacing increases, implying a decrease in critical surface density. Ultimately values of spacing and density are attained such that the critical size is infinite. For our purpose, we assume that the relationship between critical surface densities of the infinite array and those of a reflected infinite slab* of the same fissionable material depends simply upon the size of unit in an array.

The infinite uniform slab is introduced because its critical thickness is easily computed and can be related to critical surface densities of a large family of arrays. For example, a water-reflected infinite slab of U(93) metal would have a critical thickness of 0.68 in., so it could be viewed as made up of 0.68-in. cubes in contact. Larger cubes would have to be spaced for criticality, and the surface density would decrease below the slab value of 32 g/cm^2 (or 30 kg/ft^2). Results of Monte Carlo calculations indicate that this decrease will not exceed 60%** if the mass of a unit is limited to about 0.3 of a critical unreflected mass of similar shape.

The guiding Monte Carlo data are illustrated by the lower curves of Fig. 26. One of these curves, provided by J. T. Thomas, gives the ratio of surface density of a reflected U(93) metal array to that of

*Note that an infinite planar system would necessarily be well-reflected, because, unlike a finite form, it is as good a target for neutrons returned from distant objects as from nearer ones.

**The 50% decrease suggested in the report that preceded this account has been adjusted for consistency with the data by Stevenson and Odegarden which will be cited.

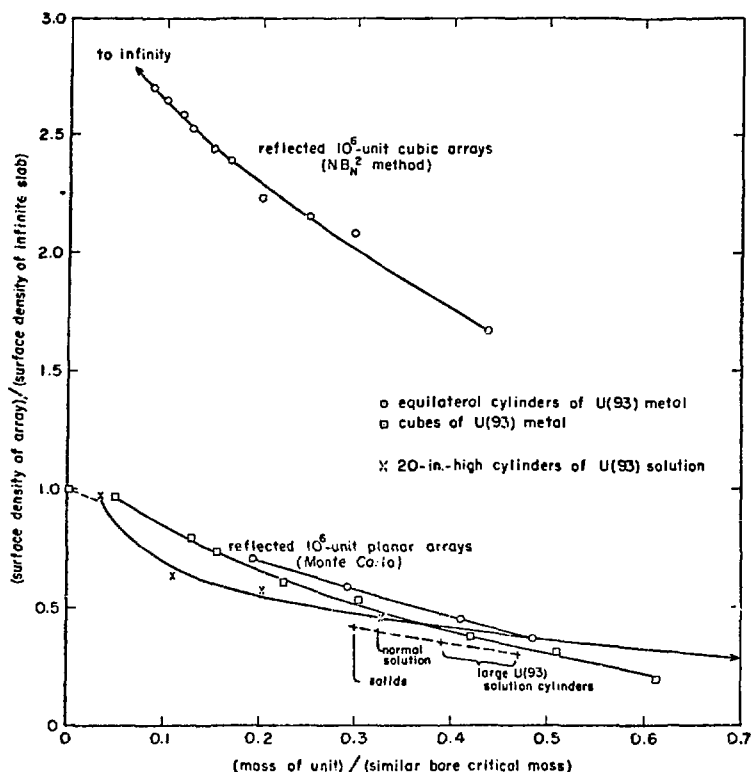


Fig. 26. Critical surface densities of air-spaced arrays relative to surface densities of critical reflected infinite slabs. Note that values for cubic arrays are much greater than those for planar arrays.

the reflected slab as a function of unit size.³⁴ The similar curve for $U(93)O_2F_2$ solutions at the atomic ratio $H/^{235}U = 44$ is adapted from data by Stevenson and Odegaard.³⁸ Reference values and unit limits from Table VII are represented by the horizontal line for solids, by the dotted extension to the abscissa 0.33 for the basic solution cylinders, and by the further dotted extension for the two larger cylinders of U(93) solution of unlimited concentration.

Although we have associated the surface-density concept with planar arrays, the values deduced are still more conservative when applied to three-dimensional arrays (which are necessarily finite). This is illustrated by the upper curve of Fig. 26, which refers to critical reflected arrays of one-million U(93) metal cylinders as established by Thomas' reliable extrapolation scheme.³¹ For "0.3-critical" units, the critical surface density is seen to exceed

the reference value from Table VII by a factor of 4. Similarly, this factor of conservatism ranges from >2 to >6 for the critical arrays listed in Table VI. (Values increase from the lower right-hand to the upper left-hand entries.) Moreover, in many practical layouts, this large margin of safety is found to be inherent.

Unlike that of a planar array, which may be characterized by an individual cell, the surface density to be considered for a cubic array is the total mass of a column of units divided by the base area of a cell. This difference implies that the two types of arrays containing mixed units cannot, in general, be handled similarly. Although the various cells of a mixed planar array can be evaluated independently, the treatment of a three-dimensional array in which units are mixed within columns and rows is not so straightforward. In such a case, one can sometimes use information about equivalent units of

different types³⁶ to construct an equivalent array in which the complication is eliminated. Otherwise, our simple rule would not apply.

GROUPS OF A FEW FISSILE UNITS

The safety of certain irregular arrays of dissimilar units can be judged by evaluating a regular array that is known to be more reactive^{*} than the actual arrangement. For example, each unit may be replaced by the largest, most reactive unit, and each spacing may be reduced to the smallest. Sometimes, however, this sort of approximation may be extremely poor, or even impossible, especially when process vessels of different shapes are clustered.

There is a time-honored means of generalizing the abundant data for several interacting units that appear in Part II of TID-7028. This generalization is based on the correlations of Fig. 27, by Henry, Knight, and Newlon, of the Oak Ridge Gaseous Diffusion Plant.³⁹ Each point of the figure applies to a critical cluster of equally reactive fissile units. The ordinate is the fractional solid angle subtended at the most nearly central unit by the other units of the cluster (i.e., the subtended solid angle divided by 4π). The abscissa is the reproduction number, k , that one unit would have if it were not influenced by the others. In other words, $1 - k$ is a measure of the interaction within the cluster. The values of k in the figure were computed by a method that the authors checked against a variety of individually critical systems.

To understand Fig. 27, we should have some idea about the relationship between a value of k and the fraction of the critical size to which it corresponds. For a fissile system that contains little poison, k is roughly the ratio of a dimension to the corresponding dimension of the critical system of the same shape. Thus $k = r/r_c$, the ratio of actual radius to critical radius, if the shape is a sphere or long cylinder, and $k = t/t_c$, the ratio of actual thickness to critical thickness, if the shape is an extended slab. (For a large, poisoned system, the ratio of dimensions will be less than the value of k .) So in terms of the fractional critical volume of a sphere, V/V_c , the value $k = 0.95$ corresponds to

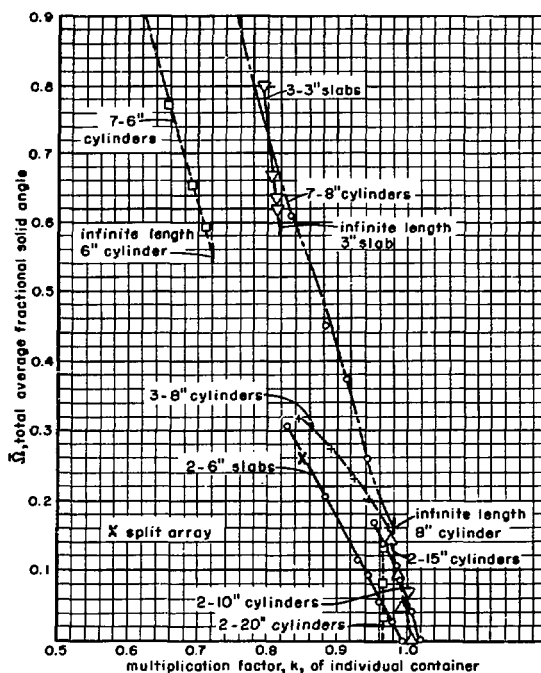


Fig. 27. Unreflected air-spaced arrays of U(93.2)-solution units; fractional solid angle subtended at central unit by other units vs computed k of an isolated unit.

$V/V_c \leq 0.86$, $k = 0.90$ corresponds to $V/V_c \leq 0.73$, and $k = 0.80$ corresponds to $V/V_c \leq 0.51$.

Now, a k of 0.9, or ~ 75% of a critical volume, is on the high side for most process equipment. Where these values are not exceeded, it appears from Fig. 27 that a subtended fractional solid angle of 0.1 or a little less can be tolerated without question. To illustrate what this means, we may consider two side-by-side cylinders of equal size whose surfaces are separated by a distance equal to the diameter. Then, according to Ref. 39, the fractional solid angle subtended at one cylinder by the other is < 0.085 . Considering that such a spacing is often inconveniently small for use in process plants, it becomes obvious that many practical interaction questions can be dismissed by inspection.

Sometimes a general upper limit to total subtended solid angle is assigned for nuclear-safety purposes. Like other limits, this has merit if recognized as somewhat arbitrary and if used sensibly. Unfortunately there have been instances where such a limit has been considered a cure-all, even to the

* One system is more reactive than another if the reproduction number, k , of the first is greater than that of the second.

point of substituting it for direct comparison with experiment. This comment, of course, is no reflection on the method, and applies equally to any formalism that is used without discrimination.

An illustration of the need for discrimination is suggested by the critical combination of solution annulus and solution cylinder shown in Fig. 7. If the diameter of the axial cylinder is about 5 in. and the inside diameter of the annulus is, say, 20 in., the combination will be safely subcritical even though the annulus surrounds the cylinder completely. A solid-angle limit, of course, would reject this configuration.

As we have implied, safety evaluation by means of solid angle is most appropriate where several large process vessels are crowded together. This is just the situation where it is pointless to attempt a precision calculation of solid angle, because interaction at small spacings is not a simple geometric concept. A judicious estimate of solid angle will do all that can be justified.

We should mention that several techniques for approximating large critical arrays make use of subtended solid angles. These include the interaction parameter method of Thomas and Scriven,⁴⁰ the interaction potential methods of Newlon,⁴¹ and an albedo (neutron reflection) scheme by Clark.⁴² Each of these methods is used with success, and the interested reader is referred to accounts by the authors.

FURTHER OBSERVATIONS ABOUT INTERACTION

Several more questions about interacting fissile units arise commonly during nuclear safety analyses. These questions and data that help to answer them will be considered now.

1. What arrangement of neighboring fissile units has the same effect as complete water reflection? For an 8-in.-diam cylinder of $U(93)O_2F_2$ solution at 538 g of ^{235}U /liter, Fig. 63 of TID-7028 gives the water-reflected critical height as 9.25 in. (the point at the extreme right). Figure 64 (TID-7028) applies to the same cylinder in unreflected air-spaced clusters. We see that a triangular cluster of three of these cylinders in contact has a slightly larger common critical height than the single water-reflected cylinder, and that a hexagonal cluster of seven of the cylinders spaced ~ 1.5 in. apart has the same critical height as the reflected cylinder. In other words, two extra similar

cylinders in contact, or six extra cylinders separated by 1.5 in., have about the same effect as water reflection.

2. Another question, about the effectiveness of cadmium (a neutron poison) between interacting cylinders, is partially answered by these same figures. Figure 63 shows that cadmium on container surfaces increases the critical height of water-flooded clusters of the cylinders, provided the surfaces are separated by several inches. Then both the effect of water as a reflector and of interaction between cylinders decreases. The unreflected air-spaced clusters (Fig. 64), however, are influenced very little by the presence of cadmium. This is consistent with the fact that high-energy neutrons, which would not be captured by cadmium, are responsible for most of the interaction among air-spaced units.

3. What is the influence of a nearby concrete wall on the critical size of an otherwise unreflected array of fissile units? The answer, for arrays of 6-in.-diam by 50-in.-high cylinders of $U(93)O_2(NO_3)_2$ solution at 384 g of ^{235}U /liter, is given by Fig. 74 of TID-7028. With spacings of 4.9 in. (12.3 cm) between container surfaces, the critical number of units in an array against an 8-in.-thick concrete wall is 9.2, as compared with 15.1 units in an array without reflection. Note that an 8-in.-thick slab of water and Plexiglas against the array is slightly less effective than the concrete, the critical number being 9.5 units at the same spacing. Roughly, then, the effect of either wall is to reduce the critical number by 40%. This influence should scale as do effects of a complete reflector about arrays of different materials (see Table V), so the fractional reduction would be greater for U(93) metal and less for dilute U(93) solution.

As the result of Monte Carlo calculations, Thomas⁴³ and Crume conclude that 5- to 6-in.-thick concrete surrounding an array is essentially equivalent to a thick water reflector. Unlike water, the concrete reflector becomes more effective as thickness is increased even beyond 10 in.

4. At least a clue about the next question, "How far apart must arrays be spaced so that their influence on each other may be neglected?" is also contained in Fig. 74 of TID-7028. The most appropriate experiment that helps answer this question is

represented by the split array at the upper left of the figure. As it seems plausible that two arrays interact much like a pair of individual units, it is appropriate to ask whether halves of the split array follow the general correlation between solid angle and k that appears in our Fig. 27. Formally, at least, the fraction of solid angle subtended by one-half of the array at the center of the other half is 0.26, and the value of k for each half if isolated from the other is estimated⁴⁴ to be 0.85.* This point happens to fall on one of the curves of Fig. 27 and so encourages us to use a solid-angle limit for judging interaction between arrays. The generally tolerable value that was discussed for individual units is a fractional subtended solid angle of ~0.1. Using this same limit, we arrive at the rule of thumb that the interaction between two arrays can be tolerated if they are separated by the largest dimension of either array. (We recall, for example, that two equal cylinders separated by one diameter fall within the suggested solid-angle limit.) Of course, any generally accepted interaction limit depends upon the safety factor allowed for each array. We assume safety margins similar to those discussed in the next section.

5. Now that we have some idea of the reduction of interaction by spacing, we ask: "What is the influence of intervening walls of various materials?" Again, Fig. 74 of TID-7028 has a partial answer for an 8-in.-thick wall of concrete, or of water and Plexiglas. We noted the array of 9.2 cylinders that was critical when simply against a concrete wall. Without change of spacing, four extra cylinders against the far side of the concrete reduce the original critical number to 8.9, and a dual array of 8.0 cylinders on each side of the concrete is just critical. As will be seen in the next section, this 13% overall effect of interaction through the concrete is a small fraction of any reasonable safety factor. At a smaller spacing, a critical array of 8.50 units

against a water-Plexiglas wall becomes 8.48 with four units against the opposite side, and 8.00 with the same number of units opposite. Note that the interaction as measured by balanced critical arrays is more extreme than would be encountered under plant conditions; the interaction with a one-half critical array on one side (four units in this case) represents a more realistic upper limit if the normal safety factors of the next section are considered.

Again, Thomas has supplemented these observations by means of Monte Carlo.⁴³ His results lead to the conclusion that 4- to 10-in.-thick concrete walls are much more transparent than similar thicknesses of water.

To the extent that arrays and slabs of fissile material interact similarly, recent measurements by McCreless, Smith, Jarvis, and Duffey¹⁸ contribute to the answer of the last question. They measured interaction through each of several materials in terms of change of critical height of a 21-in.-diam U(93) metal disk against one side of the material when a similar disk of one-half critical height was placed against the other side. Results for 8-in. thicknesses of all materials investigated and for smaller thicknesses of polyethylene are summarized in Table VIII. One of the more surprising results is the isolating effect of lead, which does not capture neutrons readily. Apparently, scattering tends to

TABLE VIII
INTERACTIONS OF TWO 21-IN.-DIAM DISKS OF U(93) METAL ON OPPOSITE SIDES OF SLABS OF VARIOUS MATERIALS^a

| Intervening Material Thickness (in.) | Single-Disk Critical Height (in.) | Interaction as Change of Critical Height (in.) |
|--|---|--|
| Air 8 | 2.81 | 0.175 |
| Concrete 8 | 2.00 | 0.04 |
| Plywood 8 | 2.24 | 0.06 |
| Beryllium 8 | 1.60 | 0.03 |
| Lead 8 | 2.22 | 0.02 |
| Polyethylene 8 | 2.16 | not detectable |
| Polyethylene 6 | 2.16 | not detectable |
| Polyethylene 4 | 2.17 | 0.05 |
| Polyethylene 2 | 2.21 | 0.40 |
| Air 2 | 2.81 | 0.67 |

*The subtended solid angle is obtained by the method of TID-7016, p. 35, where similar cells about each unit define the boundary of each half-array. The critical number of units in a single array (at the spacing of the split array) is $N_c = 13.0$ by interpolation of data from Figs. 61 and 74 of TID-7028. The value of k in each half of the split array, containing $N = 8$ units, is about the ratio of average dimension to critical dimension, so that $k = (N/N_c)^{1/3} = 0.85$.

^aOne disk is equivalent to one-half the isolated critical disk. Interaction is measured as the influence of the half-critical disk on the critical height of the other disk.

return neutrons to the plate in which they originate, and elements of high atomic weight, such as lead, are particularly effective back-scatterers. The results for concrete and polyethylene are roughly consistent with the array measurements of TID-7028 discussed above.

6. The effect of water or other hydrogenous material within an array is a combined result of moderation, reflection, and capture. At small thicknesses (less than 2 or 3 in.) or low densities, moderation and reflection predominate, and the presence of the water increases the reactivity of the array. Naturally, the effect of reflection is greater if the array is originally bare than if a reflector surrounds it. At larger thicknesses, capture predominates so that the water reduces reactivity. This complex behavior suggests the final question of this section: "What is the maximum effect of water in reflected arrays of various types of units?" There is no simple answer unless we confine our attention to reasonably large units of highly enriched uranium, such as are usually encountered in bulk storage. For 20.9-kg units of U(93) metal, the effect is demonstrated in Fig. 95 of TID-7028. Here the optimum thickness (~ 2 in.) of Plexiglas between the units of a reflected array reduces the critical average density from 1.7 to 1.0 g U/cm³, or $\bar{\rho}/\rho_0$ from 0.090 to 0.053. According to our Fig. 19, the corresponding reduction in critical mass is from 360 to 165 kg, a factor of 2.2. Less-complete data for effects of Plexiglas in arrays of U(93) solution (384 g ²³⁵U/liter) appear in Table 14 of TID-7028. The total decrease of critical array capacity caused by Plexiglas boxes around each unit is a factor of 2.1, most of which is attributed to reflection about the array. So the factor left for the effect of the boxes in a fully reflected solution array is much less than that for the metal array. (If the effect of external reflection is estimated from Table 21 of TID-7028, the remaining factor is only ~ 1.2.) An important conclusion is that the greater influence of the hydrogenous material appears in metal arrays, for which a measurement exists.

Like the preceding cases, this observation has been generalized by Monte Carlo results.³⁵ The more general conclusions, as they apply to storage arrays, are included in a storage guide discussed in the next section.

V. GENERAL CRITICALITY-CONTROL PRACTICES TOOLS FOR CRITICALITY EVALUATION

We now have a reasonable idea of the sources of criticality information and the various categories of data that are suitable for nuclear-safety evaluation. As stated before, these classes of information are: directly applicable experimental data, calculated results that are subject to verification, and semiempirical approximations that either contain factors of conservatism or also require verification in regions that are not bracketed experimentally. Ideally, the latter two categories would be used only for interpolation among experimental points. Presently, however, a number of regions of practical interest are not blanketed experimentally. Examples of deficient data include those for water-moderated plutonium of high ²⁴⁰Pu content, and those for many similarly moderated reactor-fuel mixtures such as ²³³U-Th, and Pu-²³⁵U-²³⁸U.

Where such an experimentally unknown region is encountered, the alternatives are to find more reactive systems for which data do exist, or to obtain new information. Examples of the first alternative are treating a vessel of irregular shape as a more compact cylinder or sphere of the same volume, applying the minimum critical dimension of a solution cylinder to dilute solutions, using U(93) critical data for low-enrichment uranium, assuming full water reflection instead of partial reflection, and viewing an irregular storage arrangement as a regular array with maximum planned unit size and minimum volume per unit. Usually more than one set of conservative conditions can be found, such as treating the above storage configuration first as a cubic array, then as part of an infinite planar array. If the most conservative approach does the job as well as desired, there is no need to look further.

Frequently, however, conservative application of off-specification data proves uneconomical. Then one must arrange for new data, and the help of the reactor physicist is required (of course he may also help with the first approach). He may obtain new experimental information if this is consistent with schedule and justifiable economically, or he may provide arguments for certain conservative limits derived from computation or semiempirical relations. A third choice may be to use reasonable estimates of critical conditions (instead of certainly conservative

estimates) for operational planning, followed by arrangements to check out the safety of the actual equipment before its ultimate use. Carefully controlled neutron-multiplication measurements⁴⁵ will either confirm the safety of the operation or indicate some needed modification.

Critical conditions or conservative estimates established by some means give a suitable answer to the question, "What is critical?." But there remains the more subtle question, "What can become critical?," or, to return to words used earlier, "What can go wrong with an operation and with the people performing it?." Unlike the first question, this one has no general set of best answers. It is apparent that the "best" compromise between economy and risk must depend upon details of design, operation, and organization.

We recognize, however, that a "best" nuclear-safety answer is an idealistic goal that can be approached only to the extent that expert judgment is applied. Again, this statement implies emphasis upon judgment about the reliability of an entire operation. Although paths toward the above idealistic goal cannot be described in detail, experience has led to certain nuclear-safety practices and points of view that are accepted rather generally. The rest of this section deals with existing generalizations of this sort, while recognizing their limitations.

THE GENERAL CRITICALITY SAFETY STANDARD

Widely accepted nuclear-safety criteria appear in the brief "American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors."⁴⁶ As shown by the following discussion, this product of the American Nuclear Society and the American National Standards Institute is consistent with the general principles stated at the end of Sec. I.

One of these principles, "The protection of life is more important than the protection of property," is paraphrased in the introduction of the Standard. It is further implied by the statement that criteria to be established by management may be less stringent than usual when personnel are protected by shielding.

Another principle, "General nuclear-safety guidance is superior to stereotyped rules intended for broad application," is reflected in the general nature of the Standard's requirements. For example,

the following administrative practices are called for without attempts to specify methods of implementation.

1. safety responsibility and criteria established by management,
2. process analysis that includes effects of credible abnormal conditions,
3. controlled movement of fissionable materials including labeling and posting of limits,
4. prompt investigation and correction of process deviations that affect safety,
5. frequent safety reviews to check adherence to current procedures, and
6. emergency procedures.

Similarly, margins of safety to allow for process uncertainties and accidental conditions are required but not specified numerically.

Throughout the Standard one sees the influence of the remaining principle from Sec. I: "Risk is to be recognized, and its control is to be somewhat more stringent than that which is considered good practice in nonnuclear industry." In the introduction, risk of a criticality accident is placed on a practical basis, and the need to maintain a favorable record is implied. The administrative requirements, and recommended "double-contingency principle" and control by equipment design, are intended to go beyond nonnuclear practice.

Double-Contingency Principle. The so-called double-contingency principle is generally accepted as a guide to the proper degree of protection against operational abnormalities that are improbable but still cannot be ignored. This rule calls for controls such that no single mishap can lead to a criticality accident regardless of its probability of occurrence. It is understood, further, that there should be protection against chains of related mishaps and against combinations of other abnormalities that cannot be considered improbable. Obviously, this rather subjective rule does little more than establish a point of view about criticality control—it cannot substitute for expert judgment. Experience and common sense usually provide the only basis for classifying a conceivable mishap as "likely" or "unlikely," or for ruling it out as an impractical concept. For example, leakage between a water jacket and the vessel that it surrounds may be in any one of these categories, depending upon such things as material,

type of construction, and operating temperatures. Before leaving this subject, we should add that a mistake in a record of chemical analysis is hardly ever classed as a remote possibility, because of the frequency of slipped decimal points and transcription errors.

Geometrically Favorable Equipment. Criticality control by equipment design means selecting dimensions and materials so as to preclude criticality under the variety of conditions foreseen. It is the use of such "geometrically favorable" equipment that the Standard encourages. Although continuous processes and geometrically favorable equipment are often viewed as going hand in hand, even the safety of batch operations is controlled most effectively by limiting the dimensions or capacity of containers. This is a surer type of control than relying only on the establishment of batch size by a combination of analysis and weighing or volume measurement.

While geometrically favorable equipment is a comforting means of criticality control, it should not be viewed as foolproof—a concept suggested by the misnomer "always-safe." In every practical case, there remains some degree of administrative control that cannot be eliminated. The economic penalty associated with sizing equipment for all conceivable compositions and forms of fissile material is usually intolerable. Unless this is done, however, administrative control is required to keep the wrong materials out of the system. As examples, "safe" dimensions of U(5) solution must be scaled down by about a factor of 2 to accommodate U(93) solution, and, in the extreme, another factor of 2 would be required for U(93) metal. But even if the extreme of "always-safe" geometry were attempted, the need for administrative control would still exist. Equipment must be expected to leak, lines must be expected to plug, and the material must be introduced into and removed from even ultra-safe equipment. The resulting need to handle material outside of the equipment destroys the "always-safe" concept. There is no question about the contribution of geometrically favorable equipment to nuclear safety, but it cannot eliminate the need for informed judgment.

Choice of Administrative Practices. Reasons for the administrative practices required by the Standard are highlighted by the four most serious plant accidents reviewed in Sec. I. In each case, recognized

administrative controls broke down. Procedures, that probably seemed sensible on the surface, were improvised without allowing for abnormalities that are usually considered in nuclear-safety evaluations. The fact that all of these accidents occurred in the course of unusual cleanup operations illustrates the difficulty of maintaining effective and practical procedures under extraordinary conditions. If provisions for adapting procedures to new conditions are too cumbersome, then improvisation occurs. A high degree of managerial wisdom is required to establish procedures that maintain effective criticality control while accommodating process difficulties and maintenance.

Neutron Absorbers. The Standard permits reliance upon built-in neutron absorbers provided their effectiveness is confirmed, but it cautions against similar reliance upon absorbers in solution, because of possible loss or redistribution. Tanks packed with borosilicate glass raschig rings or containing boron-stainless grids are the permanent absorbers used most commonly to provide high-capacity solution containers. The criterion states the need for administrative control to ensure that the absorber is present and properly located, and that it remains that way. As pointed out, this problem of confirming its continuing effectiveness becomes much more difficult if the absorber is in solution instead of solid form. Unless there is an unusually reliable arrangement for checking the presence of an absorber in solution, its use as a primary means of nuclear-safety control is inconsistent with the double-contingency principle. The single error of substituting the wrong reagent can lead to criticality. Therefore, soluble absorbers are ordinarily used only for secondary protection, as in an auxiliary vessel where fissile material can appear only as the result of an unlikely mishap. An exception, which is receiving increased support, is the use of soluble absorbers for primary safety control in shielded areas, particularly where an accidental excursion is not expected to be destructive. (In other words, the double-contingency principle may be relaxed under these conditions.) The greatest economic incentive for using soluble absorbers is use of large-capacity dissolvers for irradiated reactor fuel in which solid absorbers would be incompatible with solid fuel.

Subcritical Limits. The Standard calls for subcritical limits, such as mass or volume of fissile material, cylinder diameter, or slab thickness, to be based on data derived from experiments or from calculations validated by comparison with experiments. Because examples of such limits assume reflection by an unlimited thickness of water, the reader is cautioned about reflection and interaction with other fissile material that may be still more effective. We do not interpret these examples as demanding that all fissile material within a plant be subcritical if flooded by water. Some processing organizations have adopted this requirement, but presumably to allow for a multitude of uncertain conditions such as the influence of nearby structures, equipment, and personnel. Unnecessarily rigid interpretation can lead, for example, to an awkward selection of "safe-diameter" solution storage cylinders from pipe or tubing of standard sizes. Often the environment can be controlled so that standard pipe somewhat larger than the size that would be critical with full water reflection will be safe. Alternatives, such as dropping to the next smaller size, or obtaining tubing of a special size (which has been done), can be both expensive and unnecessary.

Safety Margins. The requirement that adequate but unspecified safety margins be applied to subcritical limits is anything but straightforward. Again, judgment based on experience is called for. Some organizations have selected certain absolute safety limits, the most common for individual units being 75 or 80% of the appropriate critical mass, corresponding to dimensional limits or k values of 0.90 or 0.95. It is understood that these extreme limits apply after allowance for uncertainties of data, of analysis, and of conditions that may be encountered. Very seldom are such limits actually approached in operations outside of reactors. For example, in plants with an absolute limit of 75% of the critical mass, 50% of a critical mass will be encountered only rarely. Because of the sensitivity of arrays to spacing, absolute fractional critical mass limits are not so great for arrays as for individual units; 50% is considered the extreme limit, which, again, is never really encountered. We emphasize this difference between upper limits that allow for multiple abnormalities and corresponding real limits because we feel that the latter are usually

appropriate for interaction estimates. The typical instance where there is allowance for abnormal reflection has been mentioned. If such reflection is actually encountered, its tendency is to reduce interaction to below normal. Nevertheless, certain multiple abnormalities, such as the simultaneous extraordinary moderation of several fissile units in a cluster, can lead to increased interaction. This situation, which could occur with open containers below a sprinkler system or with interconnected vessels through which the abnormality can propagate, is recognizable and should be taken into account.

While on this subject, we caution against any tendency to use an "acceptable" safety factor when a larger factor would cost no more. In other words, we recommend against "standard" safety factors (as contrasted with lower limits) even if they could be agreed upon. Advantage should be taken of any free contribution to criticality control, such as favorable arrangements of materials and equipment, that tends to make proper operation convenient and maloperation inconvenient.

SPECIALIZED STANDARDS AND CRITICALITY SAFETY GUIDES

As must be apparent, it is possible to give more detailed guidance than appears in the general criticality safety standard. The problem, however, is how to add substance without introducing arbitrary constraints, inconsistencies, or inaccuracies. The chosen solution is to share the burden of this major step among a number of work groups, each examining further standardization associated with a limited aspect of criticality safety. As of 1972, such groups have prepared three supplementary standards, have submitted advanced drafts of another standard and a guide, and are exploring six more topics for standardization.

Other Standards. One of these supplementary standards, the "American National Standard, Criticality Accident Alarm System,"⁴⁷ applies to any means of signaling evacuation in the event of a criticality accident. The emphasis is upon dependability, in particular, the avoidance of false alarms. Criteria for the design of an alarm system are outlined, and general requirements for emergency action and post-accident analysis are stated briefly.

The "American National Standard for Safety in Conducting Subcritical Neutron-Multiplication Measurements *In Situ*"⁴⁸ gives guidance for safely

confirming or establishing criticality limits where plant conditions do not offer special protection against accidental criticality. To illustrate, a series of neutron-multiplication measurements made as quantity of fissile material is increased can be extrapolated to the critical quantity, below which one can establish a clearly safe margin. The standard enumerates the equipment criteria and good practices that protect against criticality in the course of such measurements. *In situ* measurements are especially useful to confirm the safety of expensive operations with equipment and surroundings that are too complex to be reproduced in a critical-assembly facility, to be evaluated in terms of known critical systems, or to be calculated reliably.

The other supplementary American National Standard is "Use of Borosilicate-Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material."⁴⁹ As mentioned before, large-volume vessels packed with borosilicate-glass raschig rings are used extensively for storage of enriched-uranium and plutonium solutions. This standard gives specifications for raschig rings that are suitable for that purpose, defines acceptable chemical and physical environmental conditions, specifies proper procedures for packing vessels and for maintenance inspection, and gives maximum permissible concentrations of various fissile materials in solution. Instead of preserving maximum flexibility as in other cases we have discussed, this standard defines an acceptable range of conditions that has been proven by experiment and experience. For example, solutions of low-enrichment uranium are not included—neither are variation of solution concentration limits with vessel size or boron content of the glass.

Standardization Studies. Among the subjects being considered for standardization is substitution of polyvinylchloride raschig rings for glass in chemical environments that would cause the glass to deteriorate. Another, safe processing and storage criteria for uranium of low ²³⁵U enrichment, might include the storage of low-enrichment solution in vessels packed with borosilicate glass.

Other topics being explored are:

1. criticality safety limits for special applications, such as circumstances in which moderation control is inherent.

2. nuclearly safe pipe intersections for solutions of fissile materials,
3. nuclear criticality safety and control of plutonium-uranium fuel mixtures, of importance for processing fast-reactor fuel, and
4. validation of calculational methods of nuclear criticality safety.

At an advanced stage is the draft standard, "Criteria for Nuclear Criticality Safety Controls in Operations where Shielding Protects Personnel."⁵⁰ Manuals and Guides. The remaining product of a criticality safety work group, "Guide for Nuclear Criticality Safety in the Storage of Fissile Materials,"⁵¹ is to be published for trial use and comment before being submitted for adoption as a standard. This document contains subcritical limits for many water-reflected cubic arrays of spherical fissile units. Included are U(≥ 30), ²³⁵U, and ²³³U as metal, oxide, and oxide-water mixtures (H/X ≤ 20). Effects of moderation between units, displacement of units, distortion of units and cells, substitution of concrete reflector for water, and interaction of arrays through concrete are considered and evaluated for limiting cases. Although extensive, tabulations of subcritical limits do not include arrays of solution cylinders or units of low-enrichment uranium. Because the Guide is not intended to cover all aspects of practical storage arrangements, its utility is difficult to predict until after the proposed trial period.

A number of older guides and manuals specify safety limits for a variety of conditions encountered in operations with fissile materials. Perhaps the best-known of these is "The Nuclear Safety Guide,"⁴⁴ a USAEC publication revised in 1961. This Guide is out of date in that better data are available now—some of which indicate less conservatism—and more useful documents have come into being. The criticality standards and a data compilation such as TID-7028 essentially eliminate need for the old Guide.

Nuclear safety manuals combine appropriate criticality data with numerical guidance for operations in specific plants. Such manuals, for example, apply to the Gaseous Diffusion Plant⁵² and the Y-12 Plant,⁵³ both at Oak Ridge, the Savannah River Laboratory,⁵⁴ and Hanford plutonium operations.⁵⁵ Other criticality safety compilations are the "Manual of Experimental Criticality Data,"⁵⁶ of the United

Kingdom Atomic Energy Authority, the "Guide de Criticite"⁵⁷ of the French Commissariat de l'Energie Atomique, the "Handbuch zur Kritikalitat"⁵⁸ by Thomas and Weber, "Critical Parameters of Fissionable Materials Systems and Nuclear Safety"⁵⁹ (in Russian) by Dubozskiy et al., and "Criticality Safety of Nuclear Fuel"⁶⁰ (in Japanese) prepared by the Japan Atomic Energy Society.

Guides can be of considerable help with nuclear safety problems that are straightforward or have been solved for standardized operations. But they contribute little to the design of new operations where economic considerations require that the "best" solution be approached closely. In such cases, over-emphasis on guides tends to create a stereotyped image of criticality control that falls short of results that can be achieved by evaluating each major problem on its own merits.

TRANSPORT REGULATIONS

General criticality safety criteria that can be tailored to specific conditions are appropriate when those conditions remain under the control of the organization in charge of operations. But when fissile material is to be transported by a common carrier, the shipper loses this control. Consequently, more specific criteria have been adopted for the packaging and labeling of fissile material for shipment, based on tests that simulate both normal transport conditions and hypothesized accidents. Essentially uniform requirements appear in international⁶¹ and national transport regulations. In the US, packaging requirements are established by AEC regulations⁶² and an AEC Manual chapter,⁶³ and actual shipment is regulated by the Department of Transportation.⁶⁴

The following review of these transport criteria should provide a background for discussion of examples that have occurred in practice. It is a rather free interpretation of the criteria and is not expected to substitute for the actual regulations.

The transport regulations distinguish between "undamaged" and "damaged" packages. The condition of an undamaged package is established by tests that simulate the effects of dropping during handling, extremes of summer heat and winter cold, and rain. In general, packaging intended for fissile material will not be affected by these tests. The damaged package is defined by a sequence of more extreme tests for impact, fire, and flooding, which is intended to represent effects of a severe accident.

Before interaction among packages is considered, each damaged package must remain subcritical when immersed in water. For this purpose, inleakage of water is assumed unless there is a specific individual demonstration before each shipment that such leakage cannot occur. This extreme requirement is to guard against the occasional failure to close packages properly, as by omission of a gasket.

In considering permissible accumulations of packages, three general categories of transport and package condition are recognized for fissile material.

- Class I. It is not necessary to control the number of packages during transportation—in other words, the interaction among packages is so small that it can be ignored.
- Class II. The only required control is a limitation of the number of packages in a vehicle or storage location to a specified value, the "allowable number."
- Class III. The shipper has exclusive use of the vehicle and controls loading and unloading so that advantage can be taken of certain special arrangements of packages and restricted mixing with other types of package.

For Class II, which applies to transport by common carrier, a previously existing device is used to limit the number of packages of fissile material in a vehicle or storage location. In its original form, this device is the assignment of a given number of "radiation units" to each package of radioactive material on the basis of its measured external radiation. The overall radiation level is limited by DOT regulations that allow no more than 50 radiation units per vehicle or storage location (there is a concurrent limitation of no more than 10 radiation units per package). To avoid complicating life for the carrier by assigning separate numbers for criticality control, the meaning of "radiation unit" has been modified to accomplish this control as well as its original purpose of limiting total radiation. The new term "transport index" now replaces "number of radiation units." The dual control is accomplished by defining the transport index assigned to a package as the greater of the following two numbers:

- 50/(allowable number of like packages for criticality control), or
- the number of radiation units as defined before.

Note that there is no detectable radiation outside most packages of unirradiated fissile material. Then, of course, the assigned transport index is just the first of the above numbers.

We are now ready to state interaction criteria for the various classes of fissile-material shipment.

Class I. Packaging and contents are established to satisfy the following requirements.

1. Any number of undamaged packages must be subcritical in any arrangement.
2. Two-hundred-fifty damaged packages must be subcritical in any arrangement with any distribution of water that is consistent with the results of accident tests.

As for the second requirement, it is frequently difficult to show that 250 damaged packages would be subcritical without also demonstrating that an infinite number would be subcritical.

Class II. The transport index assigned to a package for criticality control is 50/(allowable number of packages) where the allowable number of packages satisfies both of the following requirements.

1. Five times the allowable number of undamaged packages are subcritical in any arrangement surrounded by the equivalent of a complete water reflector. This is supposed to be more extreme than conditions at a transfer point where the accidental combination of loads is expected to be most probable.
2. Twice the allowable number of damaged packages remains subcritical in any arrangement with any distribution of water that is consistent with the results of accident tests. This presumes that an accident combining more than two vehicle loads of fissile material is highly improbable.

In the last chapter, we observed that hydrogenous material between U(93) units of moderate size in a cubic array decreases the critical number by a factor less than 2.5. Where that limit can be accepted and packages are not affected significantly by the accident tests, it is apparent that the second of the above requirements will follow if the first is satisfied.

Class III. Special approvals by AEC and DOT are required for each type of shipment. A general requirement is that two identical loads be subcritical if brought together in a configuration expected after an accident assuming the most reactive distribution

of water. Unlike Classes I and II, influences of effective constraints such as tie-downs may be considered.

When the transport regulations were proposed, there was some fear that the severe, somewhat arbitrary, accident tests might lead to unjustifiably great economic penalties for large packages such as casks containing irradiated fuel. Although experience has uncovered some procedural difficulties, the various criteria have not proven to be serious deterrents in spite of their restrictive appearance.

CONTINUING INTEREST

Although rigid, the transport criteria are in terms of performance instead of design detail. This implies developmental problems similar to those encountered in other aspects of criticality control, but within a more restrictive framework.

Thus, in all respects, criticality safety continues to be viable and challenging. This is reflected in the lively activities of the American Nuclear Society's Nuclear Criticality Safety Division (numbering more than 200 members). Titles of special sessions at ANS national meetings, sponsored by that Division, give an idea of the diversity of interests.

- November 1968: Reviews of Recent Criticality Experiments and Plans for Obtaining Nuclear Criticality Safety Data.⁶⁵
- June 1969: Nuclear Safety—Tutorial;⁶⁶ and Nuclear Criticality Safety—Techniques, Standards, and Administration.⁶⁷
- December 1969: Criticality Problems of Synthetic Actinide Elements.⁶⁸
- June 1970: Nuclear Criticality Safety Computations—Tutorial.⁶⁹
- November 1970: Implications—ANSI Criticality Safety Standard N16.1.⁷⁰
- June 1971: International Developments in Criticality Safety.⁷¹
- October 1971: Criticality Safety in the Light-Water Reactor Fuel Cycle.⁷²
- June 1972: Education for Nuclear Criticality Safety.⁷³

The sessions emphasizing instruction were cosponsored by the ANS Education Division. There continue to be worthwhile topics.

The Nuclear Criticality Safety Division also sponsors the ANS-8 Standards Subcommittee that develops the standards discussed earlier. This activity,

too, is never-ending. Not only are new standards needed, but existing ones must be refurbished or retired each five years.

Because of the continuing need for new information, new applications, and further standards activities, there is no excuse for criticality control to degenerate into dull formalism.

ACKNOWLEDGMENTS

Valuable contributions from all members of the criticality safety community are acknowledged gratefully. The helpful hand of David R. Smith was felt throughout, William H. Roach conducted numerous special computations, Elizabeth B. Johnson provided results of experiments before publication, and Joseph T. Thomas gave generously of his comprehensive data on arrays. There were perceptive reviews and encouragement by Dixon Callihan, A. J. Clark, Jr., John A. McBride, and Robert L. Stevenson.

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APPENDIX

SELECTED FIGURES FROM TID-7028, "CRITICAL DIMENSIONS OF SYSTEMS CONTAINING U^{235} , Pu^{239} , AND U^{233} ."

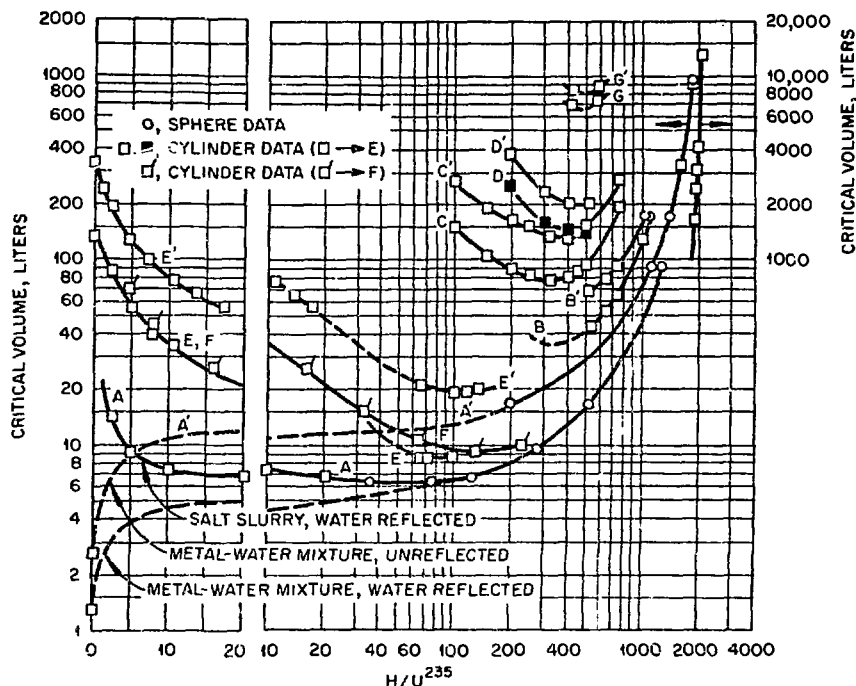


Fig. A1. Critical volume of ^{235}U -enriched uranium in spherical geometry as a function of $H/^{235}\text{U}$ atomic ratio.

Curve A: $\text{U}(93)\text{O}_2\text{F}_2$ solutions and $\text{U}(95)\text{F}_4\text{-CF}_2\text{-CH}_2$, water reflected.

Curve B: $\text{U}(4.9)\text{O}_2\text{F}_2$ solutions, water reflected.

Curve C: $\text{U}_3(4.9)\text{O}_8\text{-C}_{57}\text{H}_{110}\text{O}_6$, water reflected.

Curve D: $\text{U}(2.0)\text{F}_4\text{-C}_{25}\text{H}_{52}$, water reflected.

Curve E: $\text{U}(37)\text{O}_2\text{F}_2$ solutions and $\text{U}(37)\text{F}_4\text{-CF}_2\text{-C}_5\text{H}_8\text{O}_2$, water reflected.

Curve F: $\text{U}(29.8)\text{F}_4\text{-CF}_2\text{-CH}_2$, paraffin reflected.

Curve G: $\text{U}(1.42)\text{F}_4\text{-C}_{40}\text{H}_{81}$, water reflected.

The primed letters indicate unreflected values.

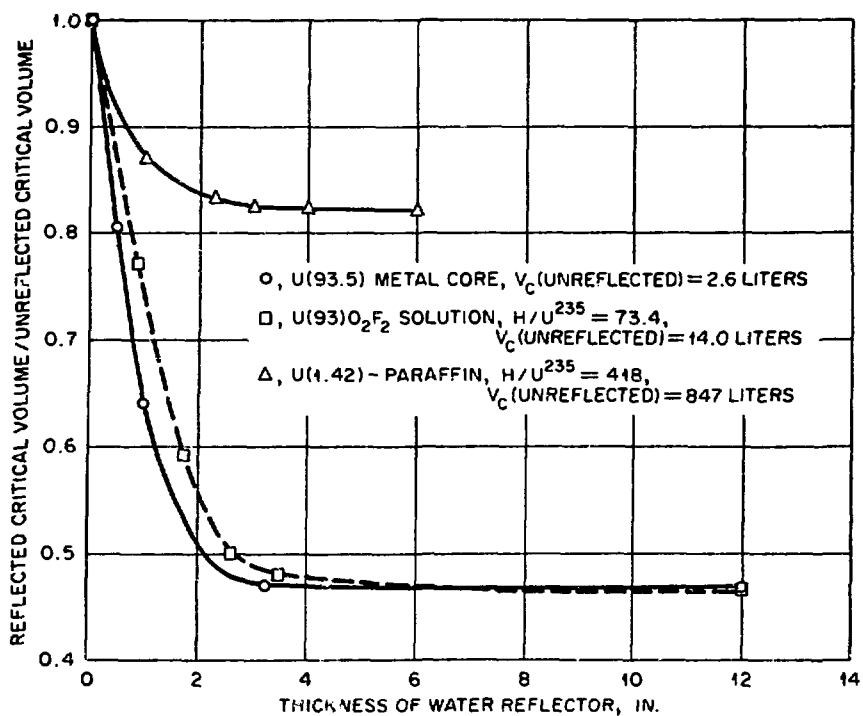


Fig. A2. Ratio of water-reflected-sphere to unreflected-sphere critical volumes vs reflector thickness.

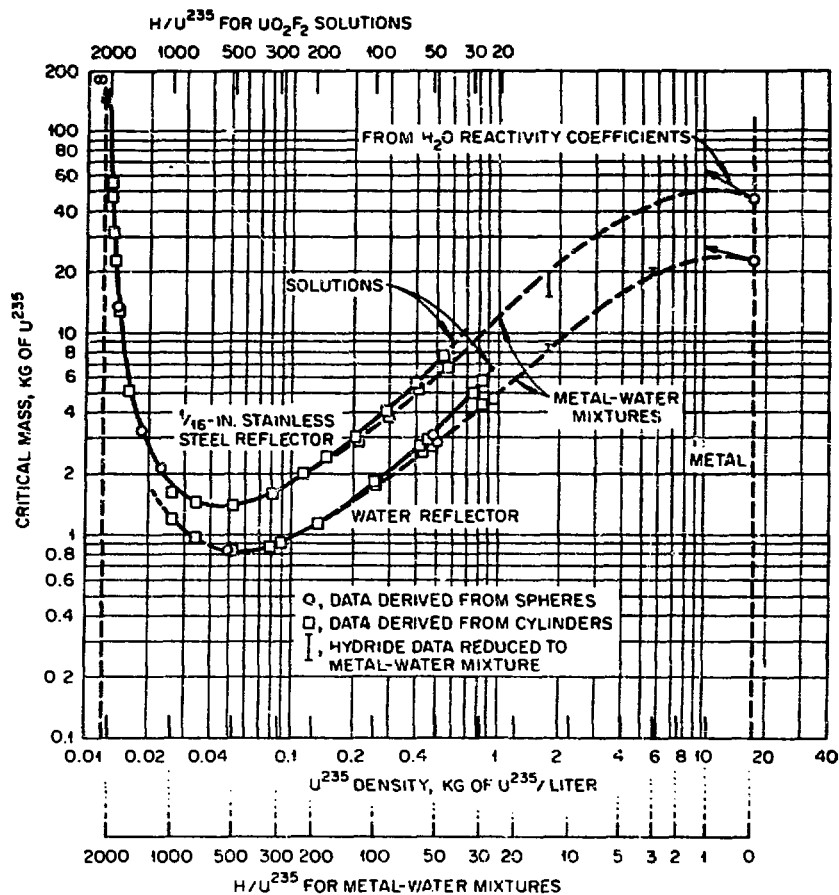


Fig. A3. Critical masses of homogeneous water-moderated U(93.2) spheres.

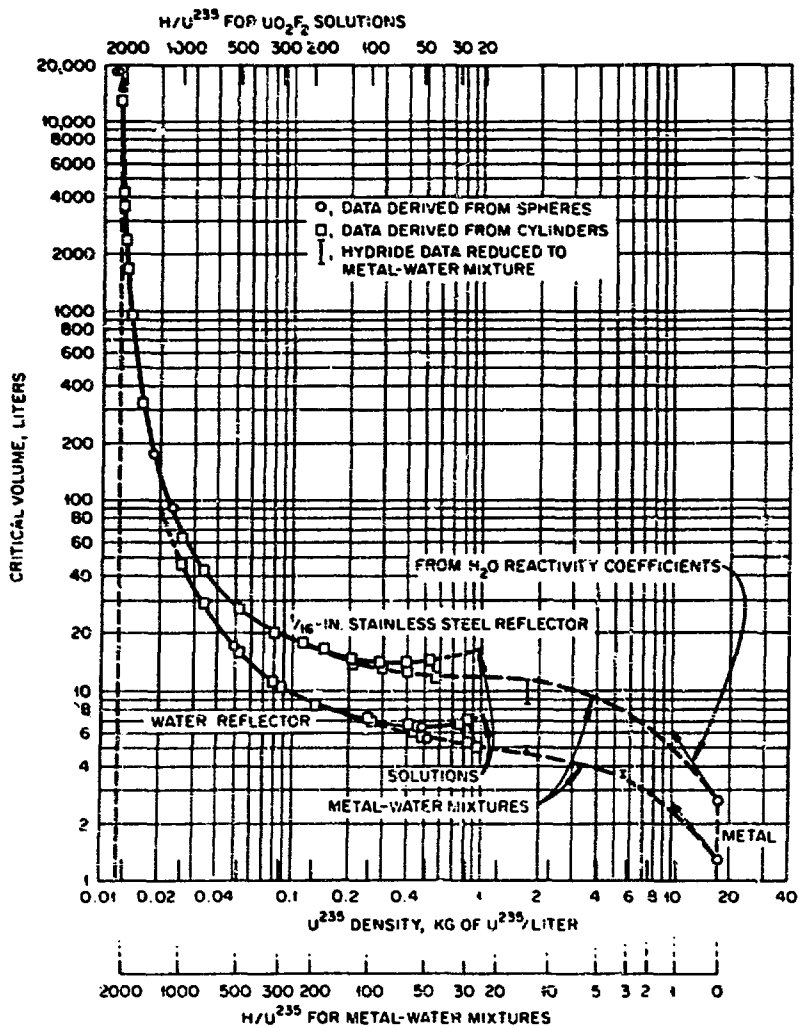


Fig. A4. Critical volumes of homogeneous water-moderated U(93.2) spheres.

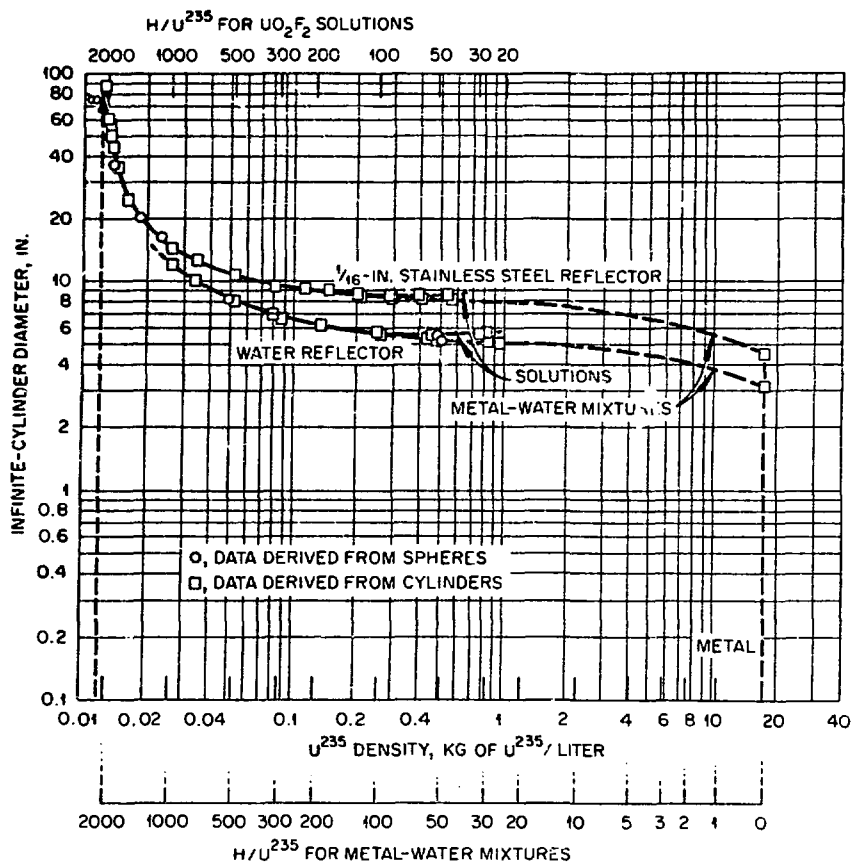


Fig. A5. Estimated critical diameters of infinite cylinders of homogeneous water-moderated U(93.2).

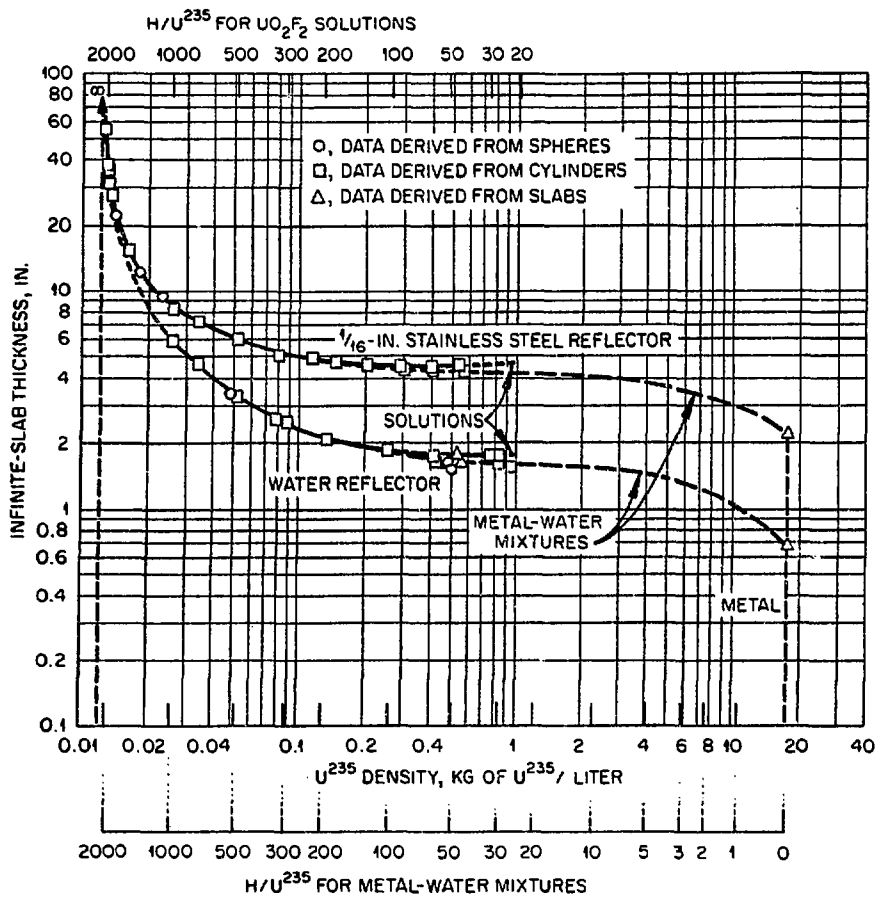


Fig. A6. Estimated critical thicknesses of infinite slabs of homogeneous water-moderated U(93.2).

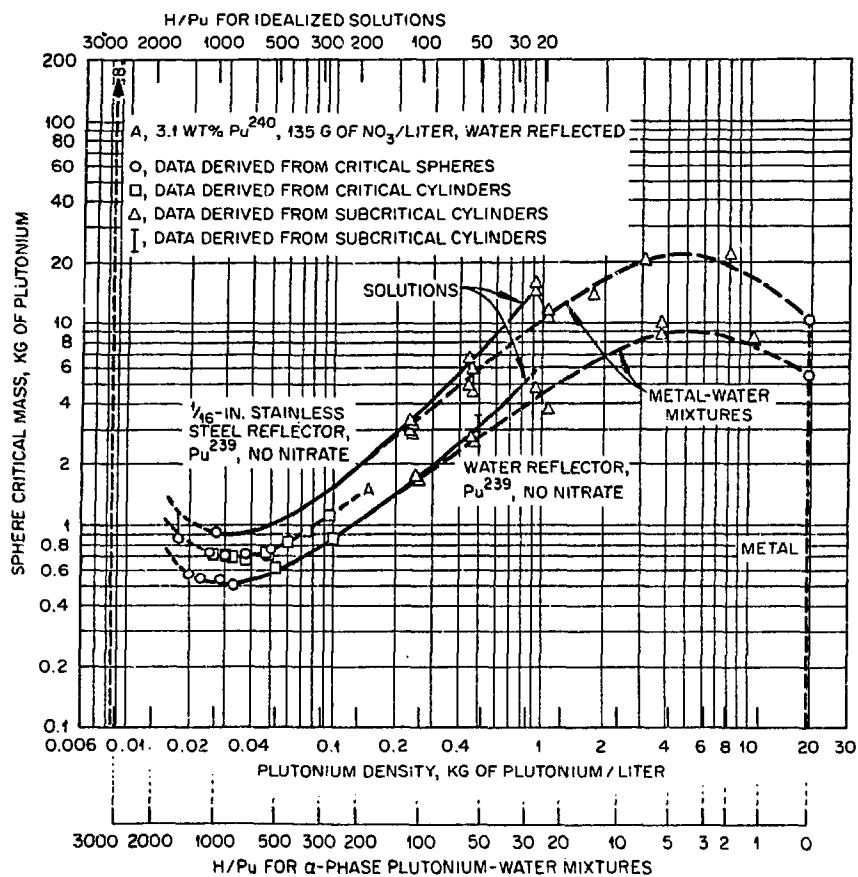


Fig. A7. Critical masses of homogeneous water-moderated plutonium spheres.

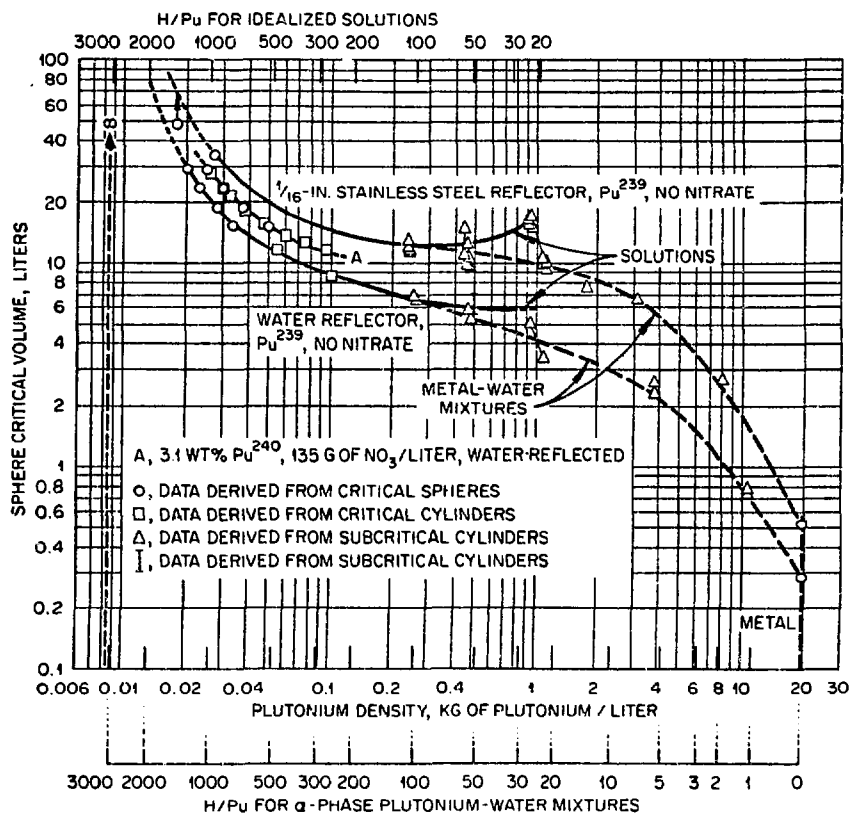


Fig. A8. Critical volumes of homogeneous water-moderated plutonium spheres.

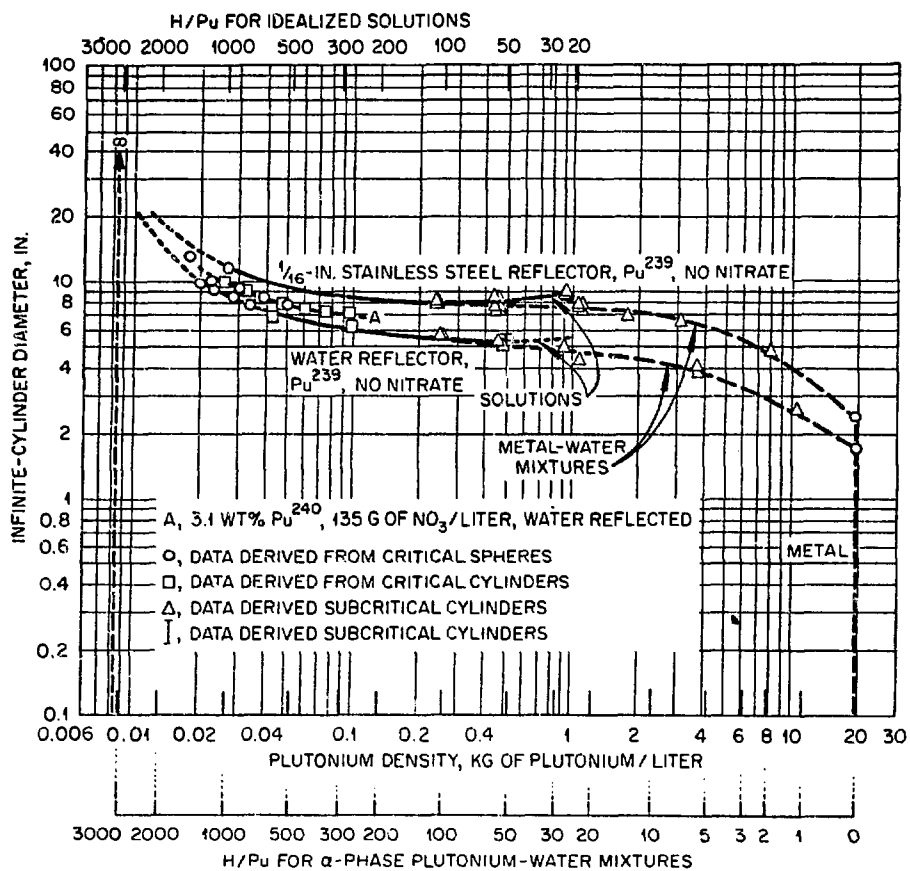


Fig. A9. Estimated critical diameters of infinite cylinders of homogeneous water-moderated plutonium.

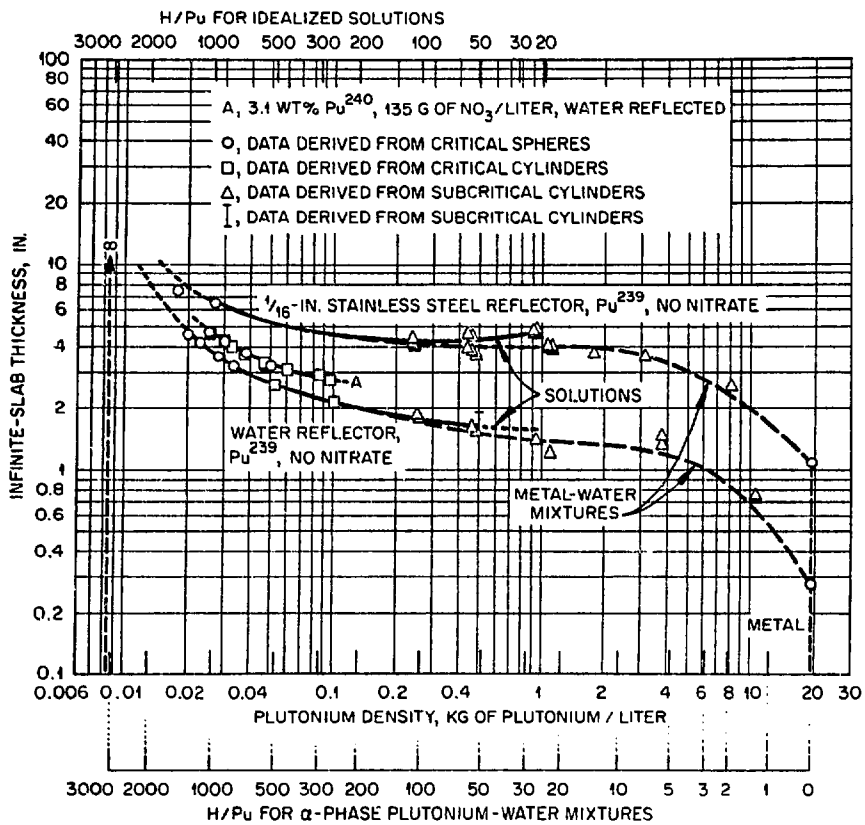


Fig. A10. Estimated critical thicknesses of infinite slabs of homogeneous water-moderated plutonium.

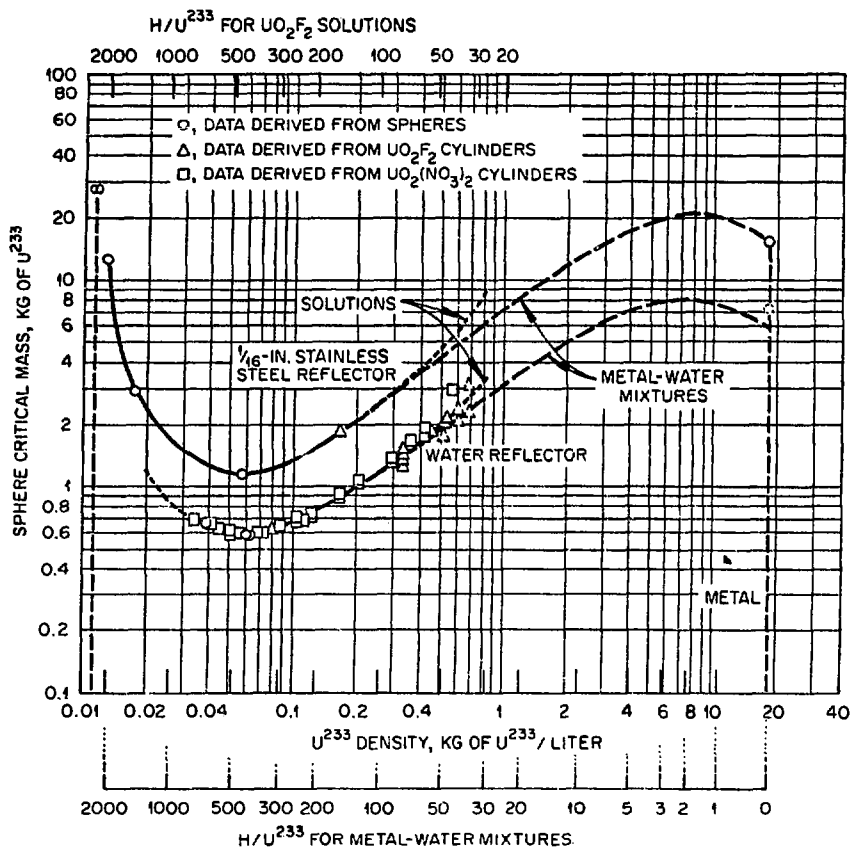


Fig. A11. Critical masses of homogeneous water-moderated ^{233}U spheres. Dashed symbols represent less certain values.

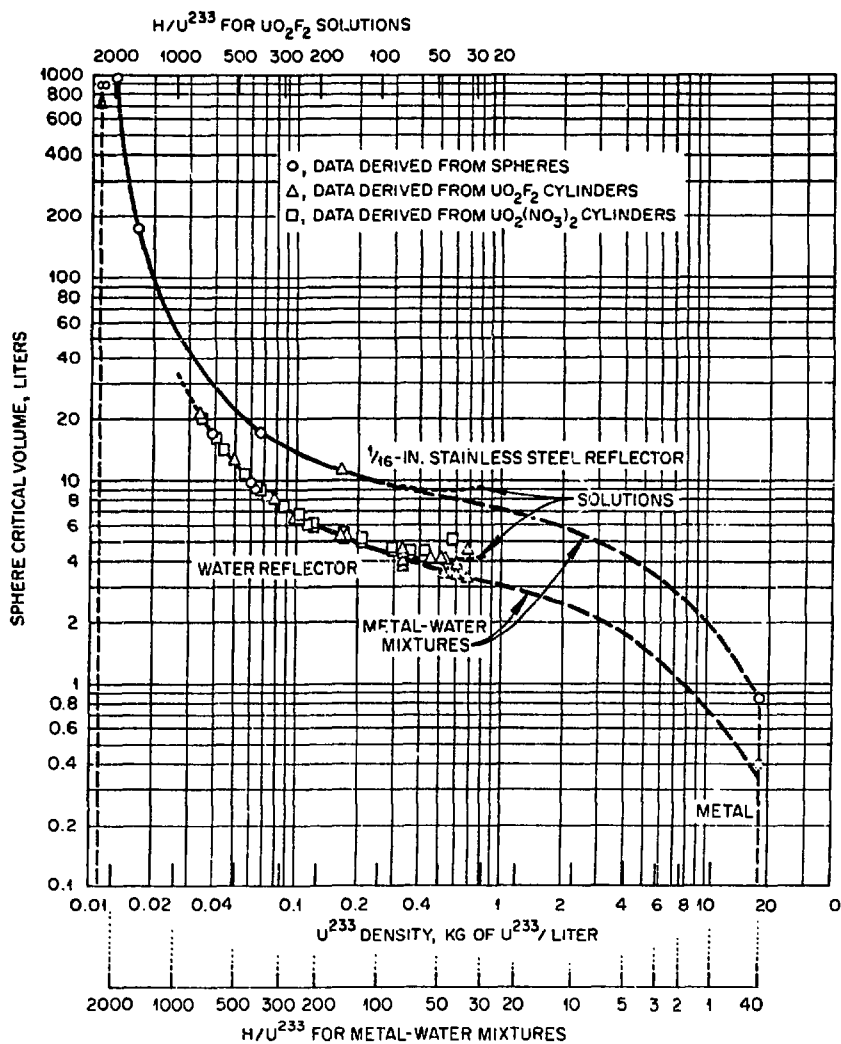


Fig. A12. Critical volumes of homogeneous water-moderated ^{233}U spheres. Dashed symbols represent less certain values.

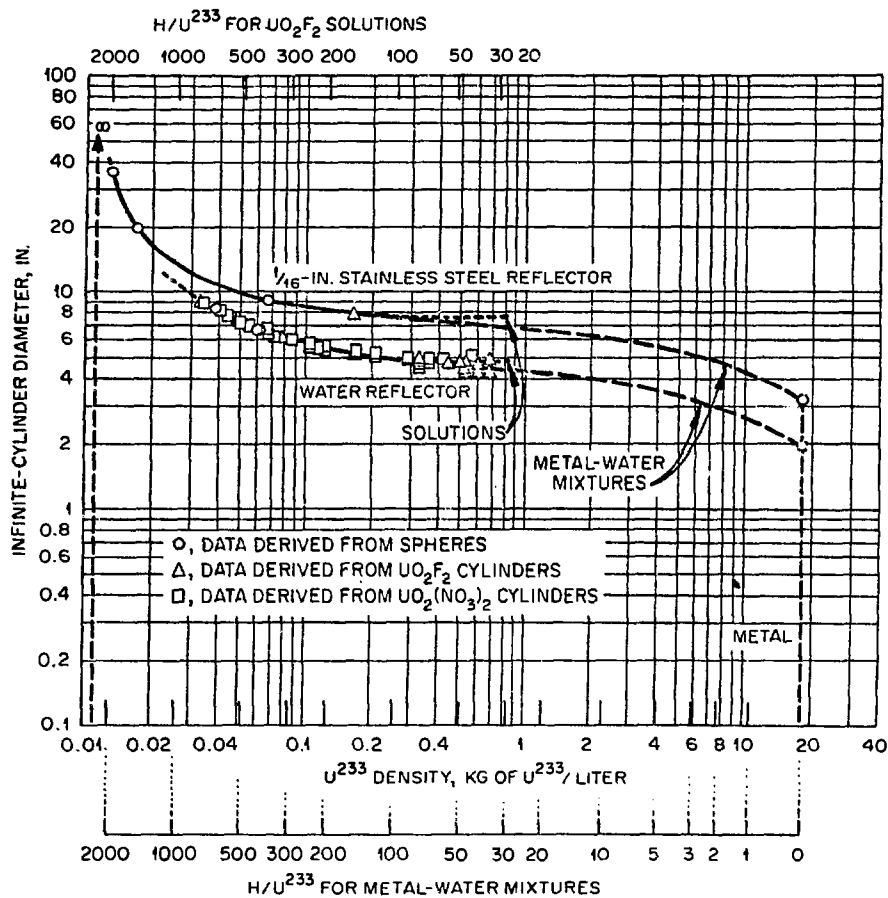


Fig. A13. Estimated critical diameters of infinite cylinders of homogeneous water-moderated ^{233}U . Dashed symbols represent less certain values.

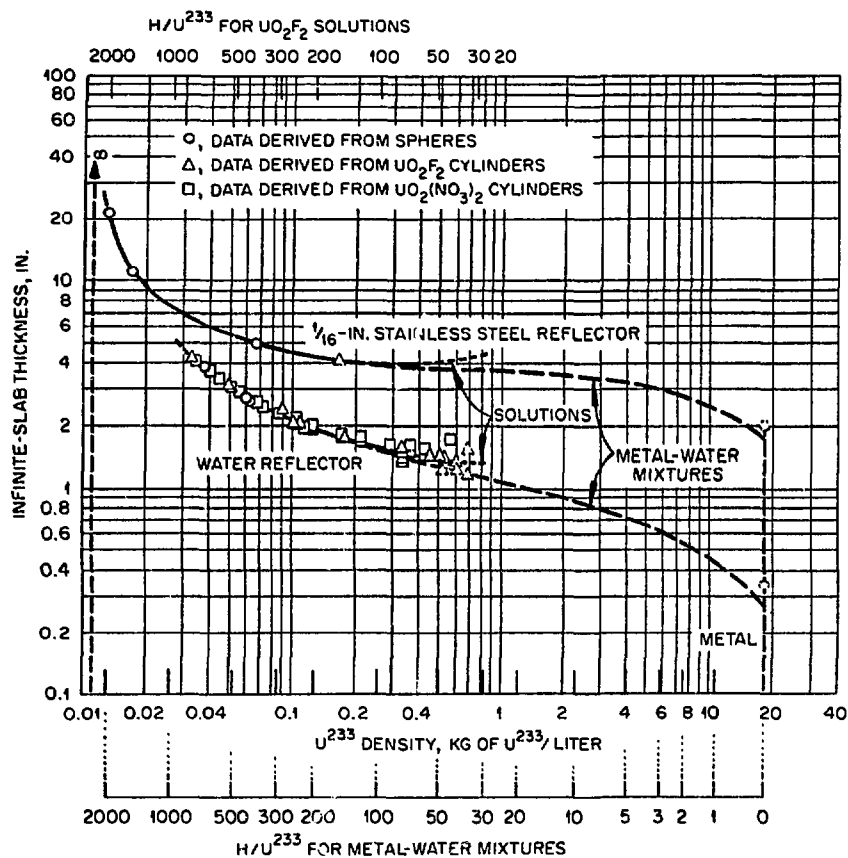


Fig. A14. Estimated critical thicknesses of infinite slabs of homogeneous water-moderated ^{233}U . Dashed symbols represent less certain values.

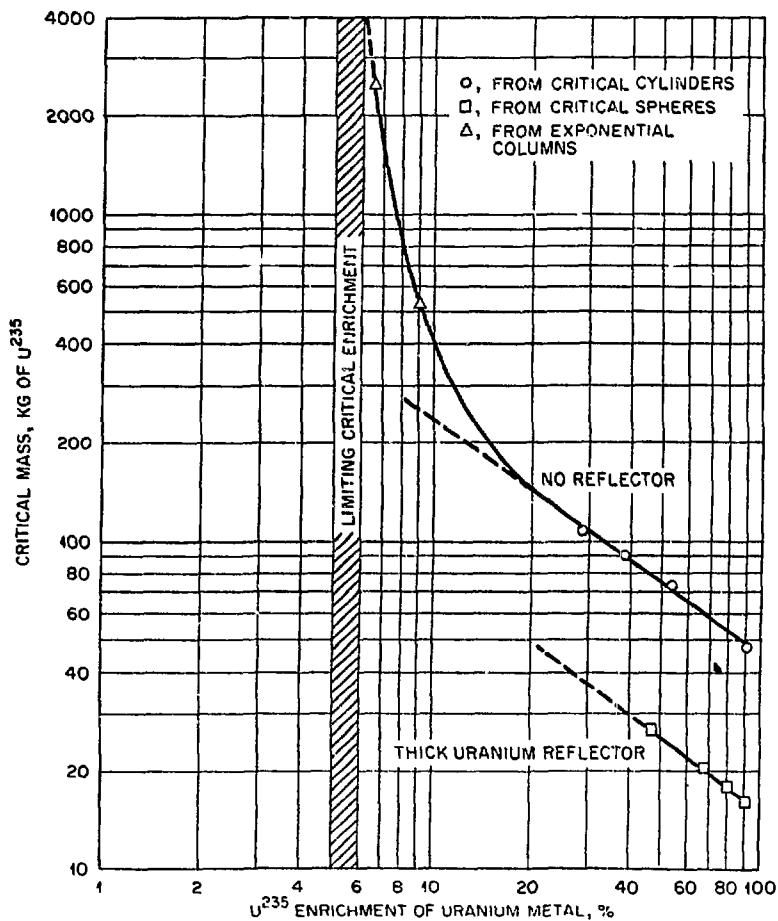


Fig. A15. Critical mass vs ^{235}U enrichment of uranium metal. The shaded strip represents the range of uncertainty in the value of ^{235}U enrichment below which uranium metal cannot be made critical.

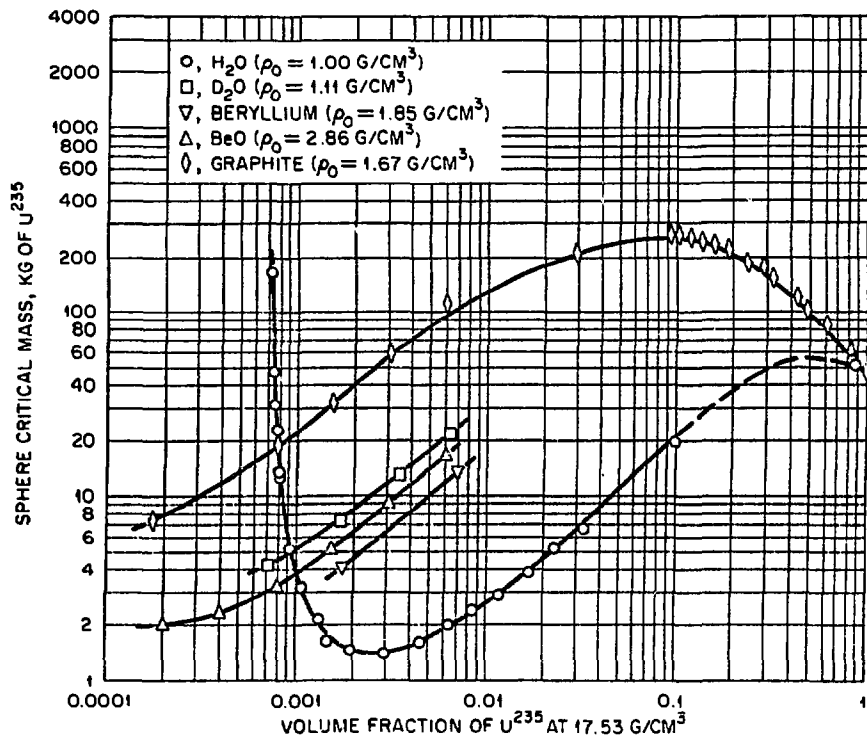


Fig. A16. Critical masses of unreflected spheres of U(93) diluted with moderating materials.

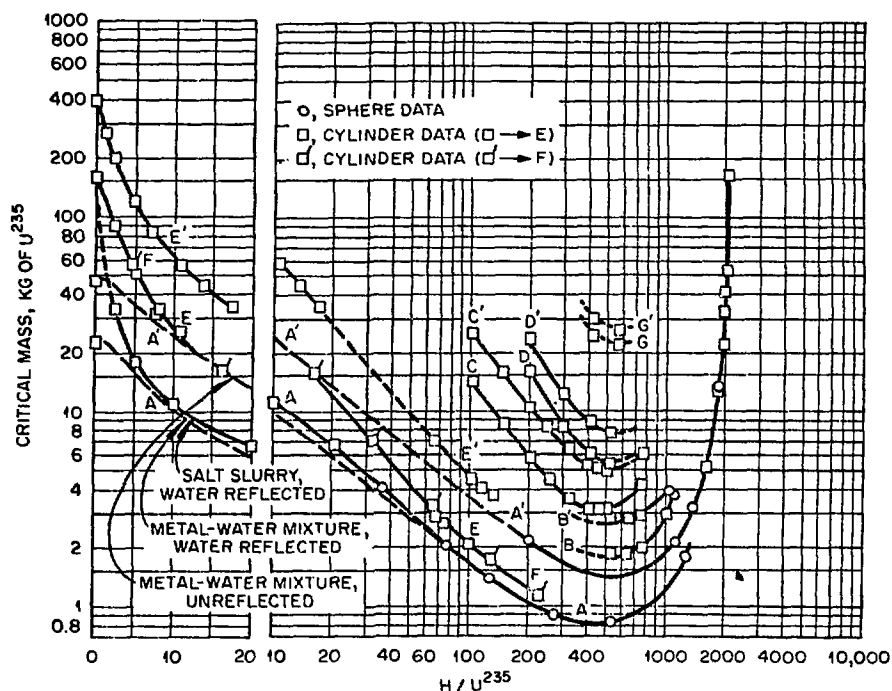


Fig. A17. Critical mass of ^{235}U -enriched uranium in spherical geometry as a function of $\text{H}/^{235}\text{U}$ atomic ratio.

Curve A: $\text{U}(93)\text{O}_2\text{F}_2$ solutions and $\text{U}(95)\text{F}_4\text{-CF}_2\text{-CH}_2$, water reflected.

Curve B: $\text{U}(4.9)\text{O}_2\text{F}_2$ solutions, water reflected.

Curve C: $\text{U}_3(4.9)\text{O}_8\text{-C}_5\text{H}_{11}\text{O}_6$, water reflected.

Curve D: $\text{U}(2.0)\text{F}_4\text{-C}_{25}\text{H}_{52}$, water reflected.

Curve E: $\text{U}(37)\text{O}_2\text{F}_2$ solutions and $\text{U}(37)\text{F}_4\text{-CF}_2\text{-C}_5\text{H}_8\text{O}_2$, water reflected.

Curve F: $\text{U}(29.8)\text{F}_4\text{-CF}_2\text{-CH}_2$, paraffin reflected.

Curve G: $\text{U}(1.42)\text{F}_4\text{-C}_{40}\text{H}_{81}$, water reflected.

The primed letters indicate unreflected values.

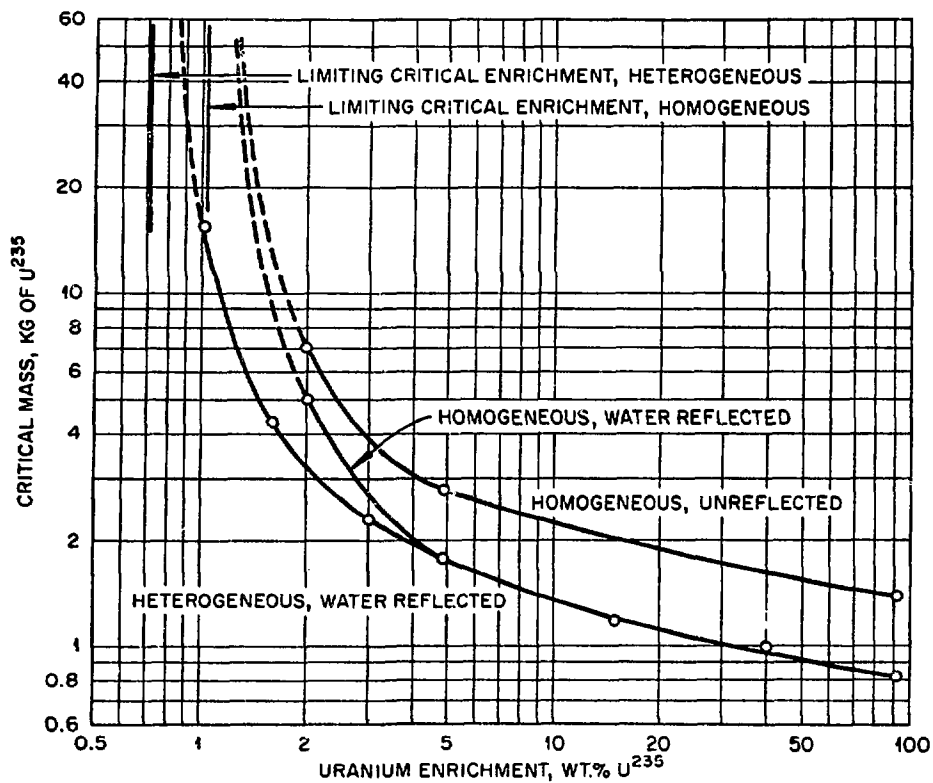


Fig. A18. Minimum critical mass as a function of ^{235}U enrichment in hydrogen-moderated systems.

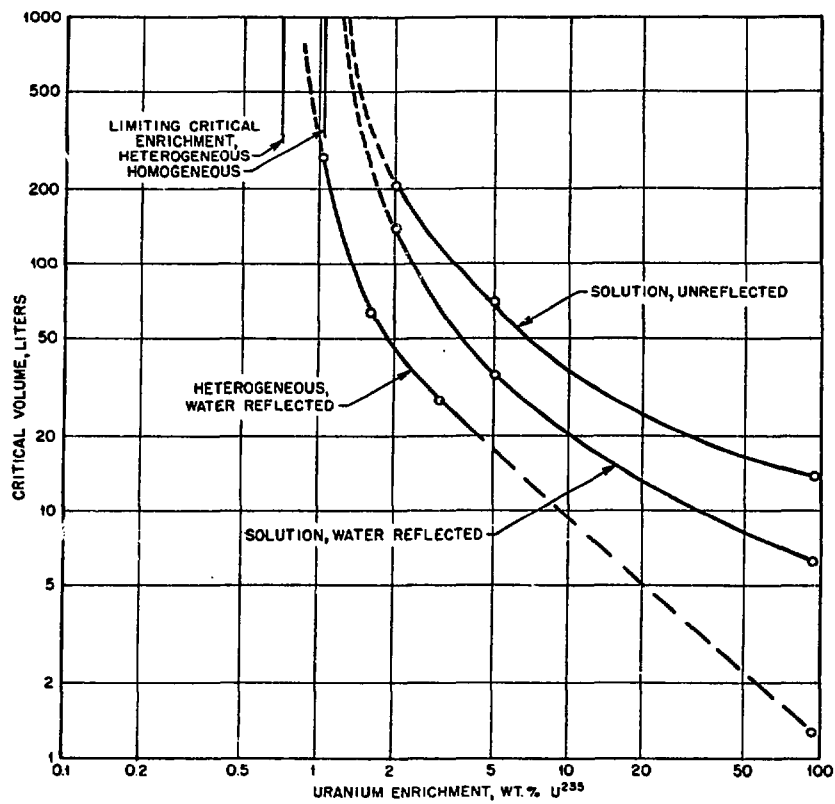


Fig. A19. Minimum critical volume as a function of ^{235}U enrichment in hydrogen-moderated systems.

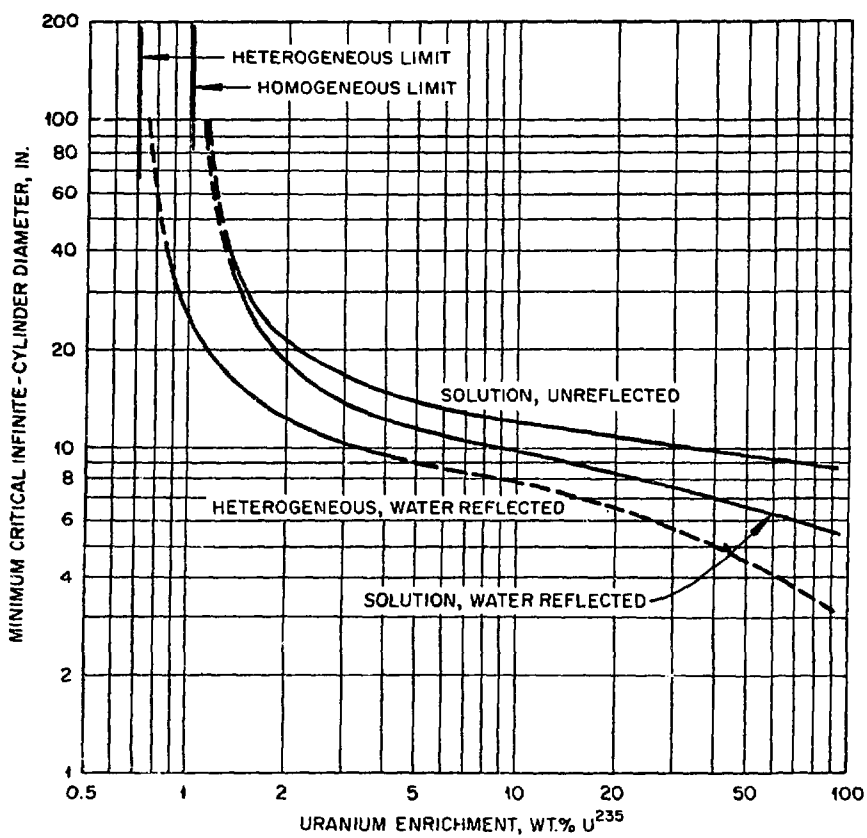


Fig. A20. Minimum critical cylinder diameter as a function of ^{235}U enrichment in hydrogen-moderated systems.

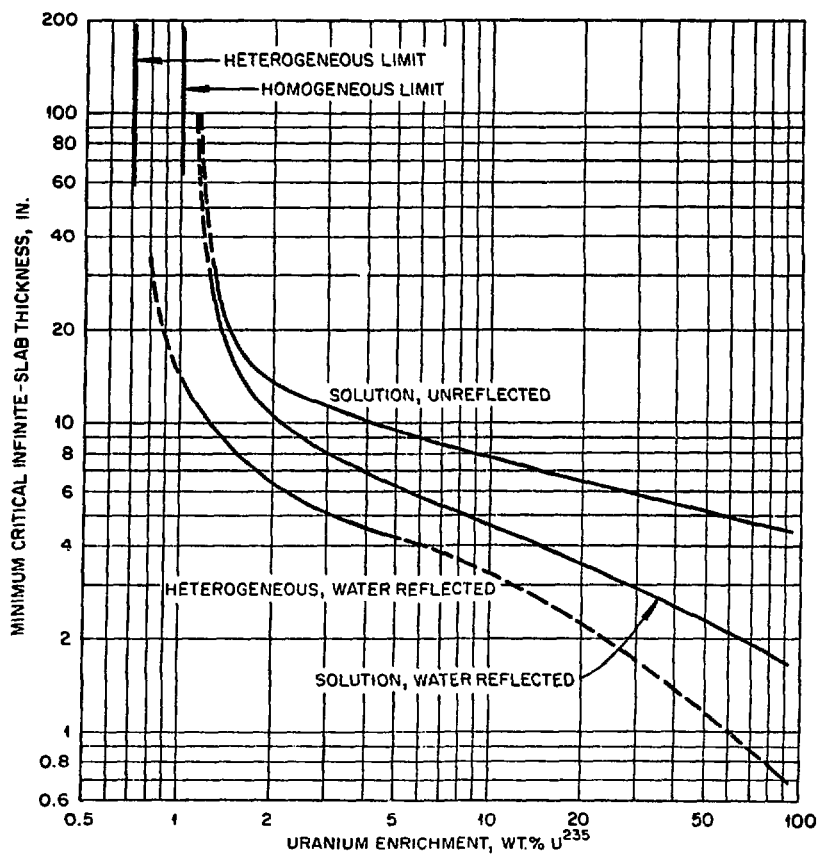


Fig. A21. Minimum critical slab thickness as a function of ^{235}U enrichment in hydrogen-moderated systems.

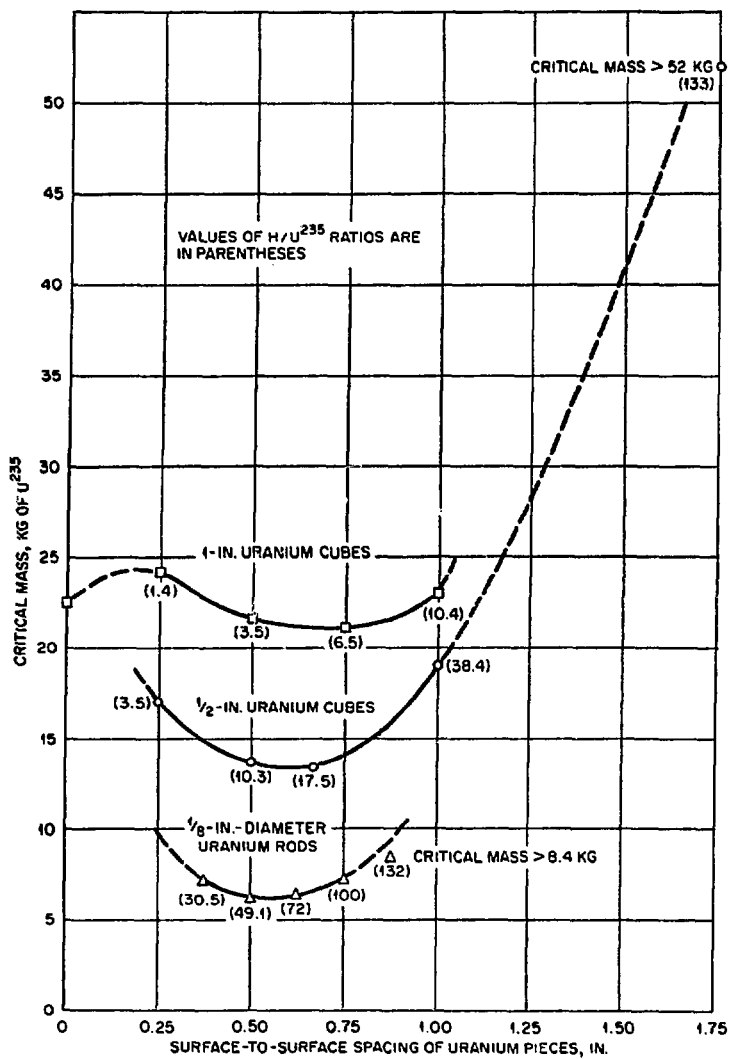


Fig. A22. Critical mass of submerged U(94) metal lattices as a function of lattice spacing.